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ABSTRACT

This handbook presents recommendations agreed upon at the meeting of the International Commission on Radiological Units and Measurements (ICRU) held in Montreux, Switzerland, in April 1962. It is written in a report form with a preface including symbols, abbreviations and definitions of terms used in the report. The report consists of four sections: (1) Direct and relative measurements of the activity of radioactive sources; (2) Low-level radioactivity in materials and its relation to radiological measurements; (3) Availability of radioactivity standards, present and future requirements; and (4) Techniques for measuring radioactivity in samples and living subjects. A list of references and two appendices are also included. (BM)

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Radioactivity

Recommendations of the International Commission on Radiological Units and Measurements

Handbook 86

United States Department of Commerce

National Bureau of Standards

SE 024 912

UNITED STATES DEPARTMENT OF COMMERCE • Luther H. Hodges, *Secretary*

NATIONAL BUREAU OF STANDARDS • A. V. Astin, *Director*

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Radioactivity

Recommendations of the International Commission on Radiological Units and Measurements (1962) (ICRU) Report 10 c

National Bureau of Standards Handbook 86

Issued November 29, 1963

(This publication supersedes parts of H78. Handbooks 84 through 89 extend and largely replace H78. For an explanation, see the Foreword. Also, for a list of these titles, see page 3 of cover.)

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Foreword

The reports of The International Commission on Radiological Units and Measurements for a number of years have been published by the National Bureau of Standards in the Handbook series. In the past, each of the triennial reports of the ICRU represented a complete restatement of the recommendations of the Commission. Because of the increasing scope of its activities, however, the Commission in 1962 decided to modify the previous practice. It will issue a series of reports presenting the current recommendations of the Commission. Each report will cover a particular portion of the area of interest to the ICRU. This procedure will facilitate revision of ICRU recommendations and also spread out in time the workload of the Commission. This Handbook is one of the new series presenting the recommendations of the Commission on one aspect of the field with which the Commission is concerned. It presents recommendations agreed upon at the meeting of the Commission held in Montreux, Switzerland, in April 1962.

The National Bureau of Standards is pleased with its continuing opportunity of increasing the usefulness of these important reports by providing the publication outlet.

A. V. Astin, *Director.*

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Preface

A. Scope

The International Commission on Radiological Units and Measurements (ICRU), since its inception in 1925, has had as its principal objective the development of internationally acceptable recommendations regarding:

(1) Quantities and units of radiation and radioactivity,

(2) Procedures suitable for the measurement and application of these quantities in clinical radiology and radiobiology,

(3) Physical data needed in the application of these procedures, the use of which tends to assure uniformity in reporting.

The Commission also considers and makes recommendations on radiation quantities, units and measurements in the field of radiation protection. In this connection, its work is carried out in close cooperation with the International Commission on Radiological Protection (ICRP).

B. Policy

The ICRU endeavors to collect and evaluate the latest data and information pertinent to the problem of radiation measurement and dosimetry and to recommend the most acceptable values for current use.

Recognizing the confusion that exists in the evaluation of different radiological equipment and materials, the ICRU is studying standard methods for the determination of characteristic data for the equipment and materials used in diagnostic and therapeutic radiology. This activity is confined to methods of measurement and does not include the standardization of radiological equipment or parts thereof.

The Commission's recommendations are kept under continual review in order to keep abreast of the rapidly expanding uses of radiation.

The ICRU feels it is the responsibility of national organizations to introduce their own detailed technical procedures for the development and maintenance of standards. However, it urges that all countries adhere as closely as possible to the internationally recommended basic concepts of radiation quantities and units.

The Commission feels its responsibility lies in developing a system of quantities and units having the widest possible range of applicability. Situations may arise from time to time when an expedient solution of a current problem may seem advisable. Generally speaking, however, the Commission feels that action based on expediency is inadvisable from a long-term viewpoint; it endeavors to base its decisions on the long-range advantages to be expected.

In 1955 the Commission entered into an official relationship with the World Health Organization (WHO). In this relationship, the ICRU will be looked to for primary guidance in matters of radiation units and measurements, and in turn WHO will undertake the worldwide dissemination of the Commission's recommendations. In 1960 the ICRU entered into consultative status with the International Atomic Energy Agency (IAEA).

The above relations with other international bodies do not affect the basic affiliation of the Commission with the International Society of Radiology (ISR).

The ICRU invites and welcomes constructive comments and suggestions regarding its recommendations and reports. These may be transmitted to the Chairman.

C. Current Program

A 2-week meeting of the ICRU was held in Montreux, Switzerland, April 2 to April 14, 1962. This meeting included the Main Commission and all of the Committees that had reports prepared for final approval. Some 70 persons attended. An additional meeting of the Commission and Committee Officers was held in Ottawa from August 21 to August 23, 1962, for the principal purposes of the preparation of the status report for the Xth International Congress of Radiology and the outlining of program objectives for the next several years.

Several meetings of committees or committee task groups have been held during the past 3 years. There were meetings of various task groups of the Committee on Standards and Measurement of Radiological Exposure—Paris in January 1961 and London in April and September 1961. The Committee on Radiobiological Dosimetry also held a meeting in April 1961. The ICRU was also represented at a meeting of the Consultative Committee on ionizing radiation of the International Committee of Weights and Measures at Sèvres in October 1961.

As noted in the last report, two joint committees had been established between the ICRU and the ICRP. The Joint Committee on RBE has met twice with ICRU participation. The Committee on Methods and Instruments for Radiation Protection has not met.

Upon the request of the United Nations Scientific Committee on the Effects of Atomic Radiations, the ICRU and the ICRP agreed to undertake a second study dealing with the Medical and Physical Parameters in Clinical Dosimetry. This committee met in New York for one week in September 1959 and for a week in Stockholm in June 1960. A report of this study entitled "Exposure of Man to Ionizing Radiation Arising from Medical Procedures with Special Reference to Radiation Induced Diseases, An Inquiry into Methods of Evaluation," was published in Physics in Medicine and Biology, **6**, No. 2, 199 (Taylor & Francis, Ltd., London, England, Oct. 1961).

Reports and recommendations of the ICRU, originally designed for medical applications, have come into common use in other fields of science, particularly where "dosimetric" considerations are involved. For this reason the committees have included in their membership some scientists having competence outside the medical radiology field. Material in the report is designed to meet physical, biological, and medical requirements wherever possible.

This has introduced a small problem in terminology. The name of the Commission includes the term "radiological". In many European countries the term "radiological" is taken as inclusive of both the physical and biological sciences. In other countries, such as the United States, "radiological" appears to carry the primary connotation of relationship to medicine. It therefore may be desirable to change the name of the Commission from "Radiological" to "Radiation." It is believed that this would be properly understood by all concerned. The question has been debated by the Commission, but final action is being delayed for future consideration.

D. The Current Series of Reports

Hitherto, the triennial reports of the ICRU have been published in single volumes. However the reports are now becoming too extensive, and in some cases too specialized, to make a single publication practicable. Beginning with this 1962 series, the ICRU reports will be issued in smaller entities, each dealing with a limited range of topics. The 1962 series extends and largely replaces the 1959 report. Revisions of the 1962 series will be undertaken individually as circumstances warrant. A full listing of ICRU recommendations, including the present series, is given on page iii of the cover of this report.

The current report series includes revision of much of the material that appeared in the 1959 report in addition to a number of new topics. The following summary indicates some of the highlights of the current report series.

Radiation Quantities and Units (Report 10a)—One of the most important changes is the revision of the section on quantities and units. This revision resulted from the thorough study by an *Ad Hoc* Committee on Quantities and Units. It includes new names for certain quantities and clarified definitions for others. It presents a system of concepts and a set of definitions which is internally consistent and yet of sufficient generality to cover present requirements and such future requirements as can be foreseen.

Physical Aspects of Irradiation (Report 10b)—This report deals broadly with the physical aspects of irradiation with a considerable amount of new material added since the 1959 report. It includes an extensive discussion of the various techniques for the measurement of absorbed dose as well as exposure. Characteristics of radiation instrumentation are covered in some detail including the more sophisticated work on standards. The section on spectra has been up-dated and a new section added on neutron measurements and standards. Available data for stopping power ratios and the average energy (W) required to produce an ion pair in a gas have been reviewed. On the basis of this review it has been necessary to modify the previous ICRU tables for these factors. This modification amounts to about 1- or 2-percent change in stopping power ratios and up to 1 percent in W.

Clinical Dosimetry (Report 10d)—Much of the Commission's work on clinical dosimetry is brought together in this report. Included is an extensive discussion of practical calibration procedures and the determination of dose along the central ray. Depth-dose data relative to stationary- and moving-field therapy have been extended as have the conversion data necessary to relate ionization measurements to absorbed dose.

The principal effort had been toward the definition of nomenclature and the indication of methods. While some examples are given and data are provided for these, in general the reader is referred to other published data. The report considers ways of increasing the accuracy and comparability in clinical dosimetry. The discussion includes not only the physical aspects of dose measurement but also the wider subject of planning treatment in such a way as to deliver the prescribed absorbed dose to a defined "target volume." It also includes comments upon the common sources of error in clinical dosimetry and discusses the information which should be recorded during treatment and that which should be reported about any new treatment technique. Appendices to this report include pertinent material taken from other reports in this series.

Radiobiological Dosimetry (Report 10e)—This report deals primarily with radiobiological dosimetry, and considers methods of improving the accuracy and intercomparability of absorbed dose measurements in radiobiology. It is in effect a handbook for the experimental radiobiologist. It emphasizes the great importance of planning the experimental work in a way which makes the dosimetry easier and more accurate and it illustrates how this can be done.

Methods of Evaluating Radiological Equipment and Materials (Report 10f)—This is the first of a new group of ICRU reports dealing with methods of evaluating radiological equipment and materials. It includes a revised discussion on the measurement of focal spots and new sections on grids, image intensifiers, and body section equipment.

E. Operating Funds

Throughout most of its existence, the ICRU has operated essentially on a voluntary basis, with the travel and operating cost being borne by

the parent organizations of the participants. (Only token assistance was available from the ISR). Recognizing the impracticability of continuing this mode of operation on an indefinite basis, operating funds were sought from various sources in addition to those supplied by the International Society of Radiology.

Prior to 1959, the principal financial assistance to the ICRU has been provided by the Rockefeller Foundation which supplied some $11,000 to make possible various meetings. In 1959 the International Society of Radiology increased its contribution to the Commission to $3,000 to cover the period until the Xth Congress. In 1960 the Rockefeller Foundation supplied an additional sum of some $4,000 making possible a meeting of the Quantity and Units Committee in 1960.

In 1960 and 1961 the World Health Organization contributed the sum of $3,000 each year to the Commission for carrying forward its work. This was increased to $4,000 in 1962. It is expected that this sum will be allocated annually, at least for the next several years. In addition, the WHO has provided substantial assistance to the Commission in providing meeting space, secretarial services, etc., for the meetings held in Geneva and Montreux.

In connection with the Commission's Joint Study with the ICRP, the United Nations allocated the sum of $10,000 for the joint use of the two Commissions for the purpose of carrying out their second study. This fund has been administered by the ICRP.

The most substantial contribution to the work of the ICRU has come from the Ford Foundation through the particular efforts of Dr. Paul Pearson. Effective in December 1960, the Ford Foundation made available to the Commission the sum of $37,000 per year for a period of 5 years. This money is to be used for such things as travel expenses to meetings, for secretarial services, and other operating expenses. To a large extent, it is because of this grant that the Commission has been able to hold the several meetings considered to be necessary to move forward actively with its program.

The International Atomic Energy Agency has allocated the sum of $6,000 per year for use by the ICRU. It is expected that this sum will be allocated annually at least for the next several years.

A valuable indirect contribution has been made by the U.S. National Bureau of Standards where the Secretariat has resided. The Bureau has provided substantial secretarial services, publication services, and traveling costs in the amount of several thousands of dollars.

The Commission wishes to express its deep appreciation to all of these and other organizations that have contributed so importantly to its work.

F. Composition of the ICRU

(a) It is of interest to note that the membership of the Commission and its committees for the period 1959–62 totals 139 persons drawn from 18 countries. This gives some indication of the extent to which the ICRU has achieved international breadth of membership within its basic selection requirement of high technical competence of individual members.

(b) The membership of the Main Commission during the preparation of this report was as follows:

Lauriston S. Taylor, Chairman	United States
L. H. Gray, Vice-chairman	United Kingdom
H. O. Wyckoff, Secretary	United States
K. K. Aglintsev	U.S.S.R.
A. Allisy	France
R. H. Chamberlain	United States
F. Ellis	United Kingdom
H. Fränz	Federal Republic of Germany
H. E. Johns	Canada
W. J. Oosterkamp	Netherlands
B. Rajewsky	Federal Republic of Germany
H. H. Rossi	United States
M. Tubiana	France

G. Composition of Committee Appointed to Draft this Report for the ICRU

W. B. Mann, Chairman, Committee I, "Standards and Measurement of Radioactivity for Radiological Use"

A. H. W. Aten, Jr.,* Vice-Chairman

R. A. Allen*	W. F. Marlow
A. P. Baerg	W. W. Meinke
R. M. Brown	G. R. Newbery
P. J. Campion	C. O. Peabody
R. A. Dudley	J. L. Putman
E. W. Emery	J. Rundo
A. P. Gandy	W. K. Sinclair*
B. Grinberg	J. Steyn
K. Hogrebe	J. Thomas
J. W. Irvine	N. G. Trott*
O. Kofoed-Hansen	H. Vetter
F. E. Leborgne	C. F. Weiss
R. Loevinger	H. M. Weiss

H. The Present Report

This report consists of four sections: 1. Direct and Relative Measurements of the Activity of Radioactive Sources, 2. Low-Level Radioactivity in Materials and its Relation to Radiological Measurements, 3. Availability of Radioactivity Standards; Present and Future Requirements, 4. Techniques for Measuring Radioactivity in Samples and Living Subjects.

The fields dealt with in sections 1, 3, and 4 were covered fairly fully in the 1959 ICRU report and the present report reviews subsequent developments.** Section 2 of the present report, however, is concerned with a field that the Commission has not previously considered, namely the problems arising from the contamination, or possible contamination, of many of the materials used in the construction of counting equipment, radiation shields, and of the chemical reagents used in the preparation of samples for radioactivity measurements. Such contamination arises not only from

*Subcommittee chairman.

** For completeness, a portion of the 1959 report is included as appendix I.

the fall-out from nuclear bomb tests; indiscriminate use of radionuclides as tracers has also contributed to the radioactive contamination of materials. A noteworthy example is the contamination of steel by Co^{60} used to indicate wear in the linings of blast furnaces.

Symbols, Abbreviations, and Definitions of Terms Used in This Report

Submultiples of the curie:
- mc = millicurie—10^{-3} curies.
- μc = microcurie—10^{-6} curies.
- nc = nanocurie—10^{-9} curies.
- pc = picocurie—10^{-12} curies.

Disintegration rates and counting rates:
- dps = disintegrations per second.
- (dps)/g = disintegrations per gram-second.
- (cpm)/ml = counts per milliliter-minute.
- (cph)/mg = counts per milligram-hour, etc.

Instruments:
- GM = Geiger-Müller, Geiger-Müller counter.
- PC = proportional counter.
- LS = liquid scintillation counter.
- PS = plastic scintillation counter.
- IC = ionization chamber.

Abbreviations:
- BEA = background equivalent activity.
- BS = bremsstrahlung.
- MDA = minimum detectable activity.
- TR = tritium ratio.

Symbols:
- ϵ = quotient of the standard deviation of S by S.
- ϵ_β = efficiency of β detecting channel.
- E_{max} = maximum beta-ray energy.
- f = counter sensitivity: quotient of counting rate by activity.
- B = background counting rate.
- N = number of counts accumulated in measuring background counting rate (equals Bt).
- N_β = observed β counting rate.
- N_γ = observed γ counting rate.
- N_c = observed coincidence counting rate.
- N_s = disintegration rate.
- N'_β = observed β counting rate corrected for background.
- N'_c = coincidence counting rate corrected for background.
- N'_γ = observed γ counting rate corrected for background.
- N''_β = observed β counting rate corrected for dead time.
- N''_γ = observed γ counting rate corrected for dead time.
- n_β = observed β-channel background counting rate.
- n_γ = observed γ-channel background counting rate.
- S = sample counting rate corrected for background.
- t = background counting time.
- T_{S+B} = combined time of counting sample and background.
- T_B = time of counting background.
- $t_{1/2}$ = half-life.
- τ = empirical parameter having the dimension of time.
- τ_D = dead time of counting channel.
- τ_R = resolving time of the coincidence unit.
- $\bar{x}$ = arithmetic mean of n results.
- x_i = result of the ith measurement in a set of n results.
- Z = atomic number.

Definitions: [1]

Accuracy	expresses the degree of agreement with the true value of the quantity being measured and is subject to the influence of unknown systematic errors.
Precision	expresses the degree of reproducibility of measurement.
Radioactive concentration [2]	is the quotient of activity by mass or by volume of the material in question.
Specific activity	is the quotient of activity by mass of the *element whose radioisotope is considered.*
Standard:	the word "standard" is used in the sense habitually accepted among those working with radioactive materials to denote a sample, calibrated with respect to activity, to be used for the calibration of further samples.
Tritium ratio (TR)	is the number of atoms of tritium in 10^{18} atoms of hydrogen (The Commission deprecates the use of the term "tritium unit" for this quantity).

[1] The text of ICRU report 10a (reproduced as appendix II of this report) contains additional definitions.

[2] This is sometimes referred to as the specific activity of the sample but is not so used in this Handbook.

International Commission on Radiological Units and Measurements (ICRU) Report 10c 1962

1. Direct and Relative Measurements of the Activity of Radioactive Sources

1.1. Introduction

It is considered necessary only to review developments in the field of absolute standardization since 1959. A portion of the contents of Appendix I, ICRU Report 1959, (National Bureau of Standards Handbook 78, pages 65–71, reproduced here as appendix I) is still relevant and this report should be considered as an extension of that document. The present report is concerned with the measurement of liquid (solution) sources from which aliquots may be taken, if necessary, to produce small samples capable of measurement by absolute counting techniques. In this chapter, no account is taken either of low-level standards requirements and techniques, or of high-level (multicurie) solid-source standardization. These latter topics could form the basis of separate reports at a later date.

1.2. Direct Measurements

There have been advances in existing techniques, concerned principally with 4π counting and with various aspects of coincidence counting (Brinkman, 1961, Gandy, 1961a; Metrology of Radionuclides, 1960; NCRP–NBS Handbook 80, 1961; Methods of Radioactive Metrology in the USSR, 1961).

1.2.1. $4\pi\alpha$ Counting

4π liquid scintillation counting of α particles has been shown to be a convenient, rapid and accurate method for standardizing solutions of α emitters (Steyn and Strelow, 1960; Seliger, 1960; Brinkman, 1961). For the method to be applicable the solution must contain no β-emitting radionuclides.

1.2.2. $4\pi\beta$ Counting

Progress in the field of $4\pi\beta$ counting has been chiefly concerned with methods of source preparation. The aim has been to reduce the self-absorption of beta particles in the source material to a minimum, since mathematical calculation of the self-absorption in a nonuniform array of crystals is very difficult and uncertain, rendering any such correction subject to some doubt.

The use of colloidal silica (uniform particles of approximately 60A in diameter) may be found advantageous in $4\pi\beta$ source preparation. The aliquot of counting solution is added to a few drops of diluted colloidal silica already in position on the mounting foil or *vice versa*. Subsequent drying under a heat lamp produces smaller crystals than are obtained from untreated sources; it is believed that the silica particles act as a nucleating or seeding agent for the crystallization process. Typical results obtained with sources prepared on polyvinylchloride-acetate copolymer (VYNS) foils in this manner show a reduction in self-absorption, over untreated, conventionally heat-dried sources, to 3.6 percent from 8.7 percent for Co^{60} (E_{max}: 0.310 Mev), to 8.5 percent from 20.3 percent for Nb^{95} (E_{max}: 0.16 Mev) and to 8.3 percent from 23 percent for S^{35} (E_{max}: 0.167 Mev) as $Li_2S^{35}O_4$ (Merritt, Taylor, and Campion, 1960a and 1960b).

Suspensions of Teflon (PTFE) used in a similar way to colloidal silica have also proven advantageous. Similarly, the use of different chemical compounds and/or addition of various wetting agents have also been used with success.

Sources of very low self-absorption have been prepared by the vacuum distillation of suitable organic compounds. This technique has been used for H^3, C^{14}, P^{32}, Co^{60}, Ni^{63}, As^{77}, Cs^{137}, and Hg^{203}. The source preparation may not be quantitative and the weight of source deposited must be determined after counting is completed, for example, by using a spectrophotometer. The self-absorption of a Co^{60} source prepared by this method, as cobalt α-nitroso-β-naphthalate, was measured to be 1.5 percent (Yaffe and Fishman, 1960).

Alternative methods of producing sources of low self-absorption include the electrochemical deposition of Fe onto gold-covered foil from a solution of $FeCl_3$ (Blanchard, Kahn, and Birkhoff, 1957); electrospraying (Carswell and Milstead, 1957); and ion-acceleration techniques. Some methods may be amenable to quantitative deposition; where feasible gamma-ray comparison measurements can be made with a quantitatively prepared source.

A further approach to self-absorption determination is the so-called "tracer technique" referred to below under coincidence counting (see section 1.2.4.).

1.2.3. 4πx-ray Counting

The determination of the number of x rays emitted by electron-capture nuclides of medium Z (within the range $Z=20$ to $Z=40$) is conveniently made in a 4π gas-filled proportional counter. At elevated pressures, the detection efficiency for x rays approaches 100 percent and a plot of x-ray counting rate *versus* pressure shows a plateau. The linear extrapolation of the logarithm of the x-ray count rate *versus* p^{-1} (the inverse of the counting gas pressure) to $p^{-1}=0$ has been shown to give too high a value for the number of x rays emitted. More reliable results have been obtained by increasing the gas pressure until a count rate "plateau" is reached (Allen, 1960).

A method has been developed to measure the x-ray source self-absorption, by using the Auger electrons as tracers, in sources which decay by electron capture and also to measure the contribution due to ejected photo-electrons from the absorbers which are subsequently used to eliminate Auger electrons (Garfinkel and Hutchinson, 1962).

It is possible to use a linear extrapolation technique by suitable choice of the coordinates; for example, the counting rate may be plotted against the filling-gas absorption appropriate to the counter. Calculation of the absorption function is somewhat simplified if the counter is spherical, since the distance from source to counter wall through the gas is then constant (Vincent, 1959; Konstantinov, 1959; Drouin and Yaffe, 1961).

X-ray counting using 4π scintillation counters has been successfully applied to the standardization of high-Z electron-capture nuclides (e.g., Cs^{131}). The x-ray absorption in NaI and CsI crystals is very high and corrections for x-ray escape introduce relatively small errors. The situation is complicated when gamma rays are also emitted from the nuclide, particularly so if some of the gamma-ray transitions are internally converted (Allen, 1960; Baronovskii and Gorodinskii, 1960).

1.2.4. Coincidence Counting

The basic principles of the coincidence technique are well known and will not be elaborated upon here. Certain specific coincidence arrangements provide very powerful techniques in the field of absolute standardization and while the principles are still simple, the corrections which need to be applied to the counting rates, to produce reliable results, merit careful consideration.

1.2.4.1. $4\pi\beta$-γ Coincidence Counting

In such a coincidence arrangement the beta detector may be a 4π gas-filled proportional counter, a 4π Geiger-Müller counter or a 4π scintillation counter. For gas-filled detectors, the preference is for a proportional counter with its lower dead time, higher permissible beta-counting rate, and consequent improved precision. The usual precautions necessary in $4\pi\beta$ counting to reduce to a minimum source-mount absorption and self-absorption should be observed, since certain corrections in the coincidence method become negligible as the beta-counting efficiency approaches unity. It is customary to employ as large as practicable sodium iodide crystals as gamma-ray detectors in order to obtain maximum overall gamma-ray efficiency.

The counting corrections applicable to $4\pi\beta$-γ measurements fall into two groups. The first group is concerned with those corrections which arise from the electronic circuitry employed in the apparatus and which are count-rate dependent, while the second group deals with the corrections pertinent to the mode of disintegration of the particular nuclide undergoing measurement.

The first group of corrections for dead-time losses, random coincidences and background count-rate has been studied by several authors. To follow the various arguments it is necessary that the nomenclature used by the various authors be very clearly understood, particularly as to what is defined as a "real" coincidence, a "random" coincidence, and an "accidental" coincidence. Table 1.1 shows the nomenclature used by some authors to describe various observed and lost coincidences. For simple decay schemes, the final equations, depending on the approximations introduced, used by various authors are:

$$N_c=\frac{N'_\beta N'_\gamma[1-\tau_R(N_\beta+N_\gamma)]}{[N'_c-2\tau_R N_\beta N_\gamma][1-N_\gamma\tau_D]}$$

[Campion, 1959]

TABLE 1.1. *Nomenclature used by various authors in calculation of coincidence corrections*

	Brinkman (1961)	Campion (1959)	Gandy (1961b)	Hayward (1961)	Wolf (1961)
Registered coincidences between successive radiations originating from one nuclear disintegration	Ordinary (real, counted)	True	Vraies	Coincidences*	
Two coincidents ($\beta\gamma_1$) and ($\beta\gamma_2$) which, due to the finite resolving time, are registered as one coincidence when γ_1 is lost in the dead time due to γ_2, β_1 being undetected, or *vice versa*, β_2 being lost and γ_1 being undetected	Real, counted	Accidental	Instrumentales-virtuelles	Coincidences*	Zufällig
Two unrelated events, one in each channel	Random	Accidental	Instrumentales-fortuites	Coincidences*	Zufällig

*Hayward considers only *registered* events in the coincidence channel, without any prefix. He corrects *ex hoc* for lost coincidences due to the mixer being dead for a time τ_D after a coincidence is registered and dead for an interval (τ_D-τ_R) after a single channel event is registered.

$$N_s = \frac{N'_\beta N'_\gamma}{N'_c}\left\{1 + N_c\tau_D + \frac{2N_\beta N_\gamma - N_c(N_\beta + N_\gamma)}{N_c}\,\tau_R\right\}$$

[NCRP–NBS Handbook 80, 1961]

$$N_s = \frac{(N''_\beta - n_\beta)(N''_\gamma - n_\gamma)[1 + N_c\tau_D - \tau_R(N''_\beta + N''_\gamma)]}{N_c[1 + \tau_D(N''_\beta + N''_\gamma)] - 2\tau_R N''_\beta N''_\gamma}$$

[Gandy, 1961]

where N_s is the disintegration rate; N_β, etc., are the observed counting rates; N'_β, etc., are the observed counting rates corrected for background; N''_β, etc., are the observed counting rates corrected for dead time; n_β and n_γ are the background counting rates; τ_R is the resolving time of the coincidence unit; and τ_D is the dead time or paralysis time of each channel (assumed equal for each channel in these examples).

These formulas are equivalent when first order approximations are made and give almost identical results for most counting systems encountered in practice (when $N_c\tau_D \ll 1$), and are adequate for most needs. The formulas are presented here as examples but should not be used without further reference to the appropriate literature. Previous formulas are discussed by Gandy (1962), by whom a new formula has been derived which includes higher order corrections and which is claimed to have a wider range of validity.

By an empirical approach, the expression for N_s may be written to a first approximation, in the form

$$N_s = \frac{N'_\beta N'_\gamma}{N'_c}\left(1 + \tau\,\frac{N'_\beta N'_\gamma}{N'_c}\right)$$

where τ is an empirical parameter having the dimension of time and which must be determined for individual radionuclides and particular equipment by an appropriate method, e.g., the two-source method using the same radionuclide and the same efficiencies, or by means of the decaying-source method (Meyer, Schmid, and Huber, 1959; Thomas, 1962).

The second group of corrections is dependent upon the particular mode of decay of the radionuclide in question. Equations may be set up to compensate for phenomena such as internal conversion of the coincident gamma ray, gamma sensitivity of the beta detector, bremsstrahlung detection in the gamma counter, and decay schemes with more than one beta branch. The correction in these cases appears as a term containing the factor $(1-\epsilon_\beta)/\epsilon_\beta$ which reduces the correction to a negligible fraction as the beta efficiency, ϵ_β, approaches unity. A combination of one or more of these corrections and the time-dependent correction is usually applied.

In those cases where electron capture occurs as one of the branches, particular care must be exercised when setting up the coincidence equations.

A variation of the $4\pi\beta$-γ coincidence method consists in determining the anti-coincidence rate between the beta and gamma channels, rather than the coincidence rate (Bryant, 1962). It is suggested that this approach provides a very useful check of the electronic apparatus when used with conventional coincidence counting. To date, no comparison between laboratories employing this method has been made.

The $4\pi\beta$-γ method has also been adapted to standardize nuclides decaying by pure beta emission, by the use of the tracer technique, $4\pi\beta$-γ(T), in which a $\beta\gamma$ nuclide is combined with the β nuclide in the same molecular structure. The beta detection efficiency, reduced by self-absorption, may be varied by adding inactive carrier to the mixed counting solution. By selecting a nuclide decaying via a suitable beta transition and coincident gamma ray, the detection efficiency for this beta energy may be determined from the ratio of the beta-gamma coincidence rate to the gamma counting rate. The beta counting rate due to the pure beta emitter in the mixture may be determined by subtraction of the known count rate of the $\beta\gamma$ emitter or by allowing the $\beta\gamma$ emitter to decay. A relationship may then be established (e.g., graphically) between the beta count rate of the pure β emitter and the beta efficiency of the $\beta\gamma$ emitter. The beta count rate may be extrapolated to correspond to 100-percent beta efficiency. Preferably the two beta energies should be equal, but provided the energies are within a factor of two, the method is satisfactory, giving an absolute standardization to an accuracy of 1 to 3 percent for the beta-emitting nuclide. The half-lives of the two nuclides must be sufficiently different to allow one to decay in order that the other may be measured alone, or the $\beta\gamma$ nuclide should be absolutely calibrated (Campion, Taylor, and Merritt, 1960).

A comparison of the activity of a S^{35} sample measured by the tracer technique (Lu^{177} $S^{35}O_4$ and as Co^{60} $S^{35}O_4$), by $4\pi\beta$ counting using colloidal silica (see above) and by differential gas counting, gave a mean result with a standard deviation of approximately ± 1 percent (Merritt, Taylor, and Campion, 1960b). A recent NPL national intercomparison of S^{35} obtained $4\pi\beta$-γ(T) results from four laboratories (3 determinations) and showed an overall spread of 3.7 percent.

1.2.4.2. γ–γ Coincidence Counting

The fast rise time of organic or plastic scintillators as gamma-ray detectors permits a reduction in the resolving time of the coincidence unit. A fast-slow coincidence arrangement using detectors of low efficiency has been successfully used to standardize Co^{60}. The output from each detector is fed to a coincidence unit of resolving time between 10^{-9} and 10^{-7} sec. Output pulses are also fed through pulse-height analyzers where the response to each gamma ray may be approximately equalized (the error introduced by mismatching is quite small, 0.3 percent, if the efficiencies differ by 10 percent). Output pulses from the analyzers are fed, together with the output

pulse from the fast coincidence circuit, to a slow ($\tau_R = 10^{-5}$ sec) triple coincidence circuit. The method thus has the advantage of short resolving time and pulse-height discrimination which reduces corrections considerably. The usual precautions must still be observed to consider angular correlation and scattered coincidences (Meyer, Schmid, and Huber, 1959). Several authors have derived expressions for the accidental coincidence rate in fast-slow systems (Bell, 1961, Shera, 1961; Mayer-Kuckuk and Neirhaus, 1960; Paul, 1960).

Where the efficiencies of the two detectors, used in γ-γ coincidence counting, are large then the expression for the sum-spectrum detection in either detector is no longer a simple term containing the product of the individual gamma-ray efficiencies (Thomas, Christensen, Fastrup, Olsen, and Villemoes, 1960).

1.2.5. Statistical Errors in Coincidence Counting

Several authors have discussed the statistical errors involved in coincidence counting (Campion and Taylor, 1961; Gandy, 1961a).

1.3. International Comparisons

1.3.1. Introduction

The number of international comparisons is less than that covered by the ICRU report of 1959 but the pattern of these comparisons is changing.

The ICRU welcomes the decision of the Bureau International des Poids et Mesures (BIPM) to assume responsibility for the distribution of materials for international comparisons, to maintain a laboratory where samples may be measured, and to correlate the results of the intercomparisons. It is intended that the distribution should be made from the BIPM to only one laboratory in each participating country. This laboratory will, in general, be the national standardizing laboratory and will, in turn, be responsible for conveying the results of these comparisons to other interested laboratories in its own country. Furthermore, the national laboratories will remain responsible for more restricted intercomparisons. The results of two international comparisons, undertaken under the auspices of the BIPM are available and are included in the review below.

Table 1.2. lists those laboratories which have taken part in comparisons together with the methods of standardization used. Table 1.3. shows the overall spread in values obtained. The NBS has distributed Na^{22} and I^{131}; the NPL: Na^{24}, P^{32}, K^{42}, and Au^{198}; AECL: P^{32} and Cs^{137}; while the international organizations IAEA and BIPM have each distributed P^{32} and I^{131}. Some comments on the individual distributions are given below. The overall spread quoted refers to the difference between the highest and lowest result reported, and is expressed as a percentage of the mean value of these two results.

1.3.2. Results of International Comparisons

Sodium 22

The 1959 NBS Na^{22} sample was measured by four laboratories using $4\pi\beta$ proportional counting, $4\pi\beta$-γ coincidence counting and by $4\pi\gamma$ ionization chambers. The results do not indicate any preference for a particular measuring technique but the overall spread of 5.4 percent may be considered unreasonable for this particular nuclide, even though the decay scheme is still subject to some uncertainty.

Sodium 24

Three laboratories measured an NPL sample of Na^{24} in March 1959. The overall spread of 3.4 percent is comparable with previous intercomparisons of this nuclide.

Phosphorus 32

Measurements of the 1959 AECL, 1960 NPL, and 1960 IAEA distributions were, with one exception, all carried out in 4π geometry and no correction for self-absorption greater than 0.7 percent is indicated. The exception was a measurement in a 2π ion chamber. The AECL distribution to 11 participants shows a surprising overall spread of 13 percent over 17 independent measurements. However, if three measurements are ignored (two $4\pi\beta$(GM) measurements and one $4\pi\beta$(PC) measurement) the overall spread is reduced to 2.4 percent. Furthermore, 12 of the 17 measurements agree to within 1.3 percent. The overall spread of the NPL distribution to four laboratories is 1.7 percent.

The IAEA distribution was measured in six laboratories; five used $4\pi\beta$(PC) counters and one used a $4\pi\beta$(GM) counter. The overall spread of the results is 1.3 percent; the overall spread of the $4\pi\beta$(PC) results is 0.7 percent. A precision of this order should be expected for an energetic β-emitter of reasonably high specific activity.

In the BIPM distribution through CEA–LMR, the overall spread of 16 results reported is 5.6 percent; 13 results lie within 2 percent. Several methods of standardization were employed; the three measurements lying outside the 2-percent overall spread were a Geiger-Müller determination, a $4\pi\beta$(PC) measurement on rather thick aluminum, and a $4\pi\beta$(PS) measurement, although other measurements using these techniques did lie within the 2-percent overall spread.

Previous comparisons have shown a 2- to 3-percent overall spread in results.

Potassium 42

One distribution of K^{42} was made, by the NPL, in 1960. Three laboratories participated and the results show an overall spread of 3.0 percent (*cf.* 2 percent in 1956–57). The source material was a solution containing 5 mg KCl per gram 10 N HCl. Eliminating two $4\pi\gamma$ ionization chamber results, the overall spread is 1.8 percent.

TABLE 1.2. *Participants in international comparisons and methods used by the laboratories*

Symbol	Name of laboratory	Laboratories from which standards were received	Methods used by laboratory in column 1
AA	Aktiebolaget Atomenergi, Stockholm, Sweden	IAEA, NBS	$4\pi\beta$(PC), LS, $4\pi\beta$(PC)γ
AECL	Atomic Energy of Canada Limited, Chalk River, Ontario, Canada.	NBS, NPL, IAEA, BIPM	$4\pi\beta$(PC), $4\pi\beta$(PC)γ, $4\pi\beta$(PC)γ(T), $4\pi\gamma$(IC)
AWRE	Atomic Weapons Research Establishment, Aldermaston, Berkshire, England.	AECL, BIPM	$4\pi\beta$(PC)
BCMN	Bureau Central de Mesures Nucléaires, Centre Commun de Recherches Nucléaires, Geel, Belgium.	BIPM	$4\pi\beta$(PC), $4\pi\beta$(LS), $4\pi\beta$(PC)γ
CEA-LMR	Commissariat à l'Energie Atomique, Laboratoire de Mesure des Radioelements, Saclay, France.	AECL, BIPM, NBS	$4\pi\beta$(GM)
CRI	Central Research Institute for Physics, Budapest, Hungary	IAEA	$\beta\gamma$
CSNI	Centro di Studi Nucleari di Ispra, Italy	IAEA	$4\pi\beta$(PC)γ
DAECRE	Danish Atomic Energy Commission, Risö Research Establishment, Roskilde, Denmark.	IAEA, BIPM, NBS	$4\pi\beta$(PC), $4\pi\beta$(PC)γ
EL	Electrotechnical Laboratory, Nagata-cho, Chiyoda-Ku, Tokyo, Japan.	BIPM	$4\pi\beta$(PC), $4\pi\beta$(PC)γ
FC	Fondation Curie, Paris, France	NBS, NPL, IAEA	$4\pi\beta$(PC)γ
GWI	The Gustaf Werner Institute for Nuclear Chemistry, University of Uppsala, Uppsala, Sweden.	AECL, BIPM	$4\pi\beta$(PC)
IAEA	International Atomic Energy Agency, Vienna, Austria	NPL, BIPM	$4\pi\beta$(PC), $4\pi\beta$(PC)γ
IAR	Institut für Angewandte Radioaktivität, Leipzig, Germany	AECL, NPL, BIPM	$4\pi\beta$(GM), $4\pi\beta$(PC), $4\pi\beta$(PC)γ, $4\pi\beta$(PS)
IBJ	Instytut Badan Jadrowych, Warsaw-Zeran, Poland	IAEA	β(GM)γ
IFM	Instituto de Fisica y Matemáticas Universidad de Chile, Santiago, Chile.	NBS, BIPM	$4\pi\beta$(PC), $4\pi\beta$(PC)γ
IKO	Instituut voor Kernphysisch Onderzoek, Amsterdam, Netherlands.	IAEA, BIPM	$\beta\gamma$, LS, $4\pi\beta$(LS), $4\pi\beta$(LS)γ, $4\pi\beta$(PS) $4\pi\beta$(PS)γ
IMM	Institut de Métrologie Mendéléev, Leningrad, U.S.S.R	BIPM	$4\pi\beta$(PC)
INP	Institute of Nuclear Physics, Czechoslovakia	IAEA	$4\pi\beta$(PC)
INSBK	Institute of Nuclear Sciences "Boris Kidrich", Belgrade, Yugoslavia.	IAEA	β(PS)γ, $4\pi\beta$, $\beta(\Omega)$
IRK	Institut für Radiumforschung und Kernphysik, Vienna, Austria.	IAEA	LS
KIA	Institutt for Atomenergi, Kjeller, Lillestrøm, Norway	NBS	LS, $4\pi\beta$(PC)
KR	Isotopen-Laboratorium der Kernreaktor Bau-und Betriebs-Gesellschaft, Karlsruhe, Germany.	AECL, NPL, BIPM, NBS	$4\pi\beta$(PC)
LNE	Laboratoire National d'Essais, Service de Radioactivité, Paris, France.	BIPM	$4\pi\beta$(GM)
NBS	National Bureau of Standards, Washington, D.C., U.S.A	AECL, NPL, IAEA, BIPM	$4\pi\beta$(PC), $4\pi\beta$(PC)γ, $4\pi\gamma$(IC)
NPL	National Physical Laboratory, Teddington, Middlesex, England.	NBS, AECL, IAEA, BIPM	$4\pi\beta$(PC), $4\pi\beta$(PC)γ, $4\pi\gamma$(IC), $2\pi\beta$(IC)
NPRL	National Physical Research Laboratory, Pretoria, South Africa.	AECL, NPL, BIPM, NBS	$4\pi\beta$(PC), $4\pi\beta$(PC)γ, $4\pi\beta$(LS), $4\pi\beta$(LS)γ
NRC	National Research Council, Ottawa, Ontario, Canada	AECL, NPL, BIPM	$4\pi\beta$(PS), $4\pi\beta$(PC), $4\pi\beta$(PC)γ
PTB	Physikalisch-Technische Bundesanstalt, Braunschweig, Germany.	NPL, AECL, IAEA, BIPM, NBS.	$4\pi\beta$(PC), $4\pi\beta$(PC)γ, $4\pi\gamma$(IC)
RCH	Royal Cancer Hospital, London, England	NPL	$4\pi\beta$(GM), $4\pi\beta$(LS)
WRL	Wantage Research Laboratory, Atomic Energy Research Establishment, Wantage, Berkshire, England.	IAEA, NBS, NPL	$4\pi\beta$(GM), $4\pi\beta$(PC), $4\pi\beta$(PC)γ

Abbreviations:
(GM) Geiger-Müller counter
(IC) Ionization chamber
(LS) Liquid scintillation counter
(PC) Proportional counter
(PS) Plastic scintillation counter
(T) Tracer technique
(Ω) Defined solid angle

TABLE 1.3. *Summary of past comparisons*

Radionuclide	Date of comparison	Issuing laboratory	Participating laboratories	Overall spread in values* percent
^{24}Na	1959	NBS	AECL, FC, NBS, NPL	(5.4)
^{24}Na	1959	NPL	NPL, PTB, RCH	(3.4)
^{32}P	1959	AECL	AECL, AWRE, CEA-LMR, GWI, IAR, KR, NBS, NPL, NPRL, NRC, PTB	2.4 (13)
	1960	NPL	IAR, NPL, PTB, WRL	(1.7)
	1960	IAEA	AECL, CEA-LMR, FC, IAEA, NBS, PTB	0.7 (1.3)
	1961	BIPM (through CEA-LMR)	AECL, AWRE, BCMN, CEA-LMR, IAEA, IAR, IFM, IKO, IMM, KR, LNE, NBS, NPL, NPRL, NRC, PTB.	1.9 (5.6)
^{42}K	1960	NPL	AECL, NBS, NPL	1.8 (3.0)
^{131}I	1959	NBS	AA, AECL, CEA-LMR, DAECRE, IFM, KIA, KR, NBS, NPRL, PTB, WRL	5.1 (19)
	1960	IAEA	AA, AECL, CRI, CSNI, DAECRE, FC, IAEA, IBJ, IKO, INS, INSBK, IRK, NBS, NPL, PTB, WRL.	6.0 (12)
	1961	BIPM (through NRC)	AECL, AWRE, BOMN, CEA-LMR, DAECRE, EL, OWI, IAEA, IAR, IFM, IKO, KIA, KR, LNE, NBS, NPL, NPRL, NRC, PTB.	1.5 (3.9)
^{137}Cs-^{137}Ba	1959	AECL	AECL, AWRE, CEA-LMR, GWI, IAR, KR, NBS, NPL, NPRL, NRC, PTB	3.1 (7.5)
^{198}Au	1960	NPL	AECL, FC, IAEA, IAR, KR, NBS, NPL, NPRL, NRC, PTB	3.1 (12)

*The value in parenthesis is the overall spread of all reported results; the other figure indicates the spread of the more consistent results.

Iodine 131

Three I^{131} comparisons (1959, 1960, and 1961) have been reported. In each case the overall spread is greater than in previous years. This may well be due to many more participants and to an increase in the number of techniques employed. The results of the NBS distribution in 1959 show an overall spread of 19 percent but contain one group having very low results. If these and the results from one other laboratory are rejected, the overall spread is 5.1 percent. An analysis of the methods of measurement shows the following mean values:

$4\pi\beta$-γ: 6.343×10^5 (dps)/ml, overall spread 3.2 percent
$4\pi\beta$(GM): 6.249×10^5 (dps)/ml, overall spread 5.1 percent
$4\pi\beta$(PC): 6.192×10^5 (dps)/ml, overall spread 4.5 percent

The IAEA distribution shows an overall spread of 12 percent, which is reduced to 6 percent if two results are rejected. The $4\pi\beta$-γ results are approximately 1-percent lower than the mean, unlike the results above, and show an overall spread of 4 percent. All results referring to the first day of measurement show an overall spread of 8.6 percent, those for the second day 7.2 percent, and for the last day 17 percent.

The BIPM distribution through the NRC, Ottawa shows an overall spread in results of 3.9 percent. Twelve of the participants show agreement within 1.5 percent. The coincidence measurements show an overall spread of 3.6 percent.

Cesium 137–barium 137

An AECL distribution in 1959 to 11 participants (13 independent measurements) shows an overall spread of 7.5 percent; 10 of the 13 independent measurements agree to within 3.1 percent.

Gold 198

One distribution by the NPL in 1960 was sent to 10 participants. Rejecting one $4\pi\beta$-γ result, the overall spread is 3.1 percent. $4\pi\beta$ measurements, omitting any correction for self-absorption, form a group distinct from $4\pi\beta$-γ measurements. The overall spread of most results within each group is 1.5-percent and the mean value of $4\pi\beta$ results is approximately 1.5-percent lower than that for the $4\pi\beta$-γ results. Despite this, the spread is less than in the majority of previous comparisons.

1.3.3. General Comments on International Comparisons

The presentation of the results by participating laboratories is most important. The corrections applied to the various measurements and the derivation of quoted errors need to be clearly stated particularly when a correlation of all results is to be made. In an attempt to achieve uniformity of presentation, a result sheet is suggested; a form of this type is being tried out by the BIPM in its current international comparisons. The layout of the suggested form is shown as table 1.4.

The overall spread of the results, even by one method, is usually in excess of the quoted precision of the method. Laboratories should conduct their own reproducibility experiments for each process in a standardization; i.e., diluting, aliquoting, transfer of activity, voltage plateau for individual sources, etc.

The ICRU has considered the problem of specifying the decay scheme and half-life of radionuclides distributed for international comparisons. In some cases there are significant differences in the literature of values of these characteristics. The results of intercomparisons are sometimes difficult to analyze when measuring laboratories use different values. While it is recognized that the adoption of specified values for intercomparison purposes may lead to systematic errors between different laboratories or different methods in one laboratory depending on individual circumstances, it is felt that this may prove preferable to the variations which will occur when laboratories choose their own values. In the case of the half-life value there are two methods to reduce such errors, viz: (a) the measurements can be made sufficiently close to the reference time that any error is insignificant, or (b) the measurements can be distributed symmetrically in time about the reference time and averaged.

When comparisons were being made by a few laboratories, the overall spread, expressed as a percentage of the mean value, i.e., the difference between the largest and smallest value in percent of the mean, was indicative of the agreement obtained. Now that many laboratories participate in comparisons, more information can possibly be obtained regarding the accuracy and we therefore recommend that the agreement of the measurements of each comparison be expressed by two numbers, the root mean square deviation in percent of the mean and the overall spread as defined above.

Results of previous international comparisons, which can be analyzed in this way, should be lodged with the BIPM in order to facilitate future examination of the progress in such comparisons.

Impurities in samples are troublesome and every effort must be made to reduce these to a minimum. Insofar as international comparisons are concerned, the presence of certain impurities is not so serious since the measurements are performed at the same time in each laboratory. Typical impurities which must be considered are K^{42} in Na^{24} and *vice versa*, P^{33} in P^{32}, and Cs^{134} in Cs^{137}.

Distributions of short-lived radionuclides (e.g., In^{116m}) present problems of transport and accurate timing. An international comparison of these is

TABLE 1.4. Suggested form for results of sample measurement

(First page) *International comparison of*

Distributing Laboratory ———— Measuring Laboratory ———— Ampoule No(s). ————

Results Reference date ————

Date of measurement	Method	Number of sources	Mean activity at 1500 GMT on ref. date, (dps)/g	Percent standard error of mean	Errors typical of individual source measurement (%)			Mean value for each dilution	Other comments
					Random		Systematic		
					Counting	Weighing aliquoting etc.			
	a	b	c	d	e	f	g	h	i

Corrections Applied (%)

Method	Decay $t_{\frac{1}{2}}$	Background	Dead time τ_s	Accidental coincidences due to τ_R	Absorption		Other corrections
					Foil	Self	
a	j	k	l	m	n	o	p

(Second page)

Preparation of Sources

Method	
Diluent and dilution factor	
Nature and thickness of supporting foil	
Nature and thickness of foil coating	
Method used for source deposition	
Method of source drying	
Weight, diameter and character of source	
Liquid scintillator composition	
Liquid scintillator volume	

Please read users' notes carefully.

not a practicable proposition at the present time and users of such nuclides, who do not wish to employ "simulated standards", would be well advised to install equipment for absolute measurement ($4\pi\beta$ counters) or to purchase standard ionization chambers which could be calibrated for the short-lived materials in a standardizing laboratory.

1.3.4. Use of the ICRU Form for Reporting Results of International Comparisons

The objective of this form is to gather together the necessary data from the participating laboratories in order that a critical and useful analysis of the results may be made. Laboratories should feel free to quote errors which they genuinely believe to be applicable. While it is intended that *all* sources of error should be considered, the practice of making arbitrary overall estimates of error is discouraged.

In general it is intended, that each measuring laboratory will submit only one form for each comparison.

Notes on Column Entries

Part 1, Results

(a) *Method.* The counting technique which has been employed should be stated, e.g., $4\pi\beta$(GM), $4\pi\beta$(PC), etc.

(b) *Number of Sources Used in Calculating the Mean.* This figure should be the total number of sources measured by any one method. If sets of sources have been prepared (via different dilutions for example), the number in individual sets should be given, e.g., 7 from dilution 1, 10 from dilution 2.

(c) *Mean Activity at 1500* GMT *on Reference Date in (dps)/g.* The activity should be corrected to 1500 GMT on the reference date and expressed

in disintegrations per gram-second of solution. Each mean value should be for *one* method after all corrections have been applied.

(d) *Percent Standard Error of the Mean Activity.* For a single set of observations, this is defined as

$$\frac{100}{\bar{x}}\left\{\frac{\sum_{i=1}^{i=n}(x_i-\bar{x})^2}{n(n-1)}\right\}^{1/2}$$

where $\bar{x}$ is the arithmetic mean obtained from n results and $\bar{x}_i$ is the result of the ith measurement in the set of n. Where more than one set of observations has been made by any one method (e.g., from several dilutions) the final figure for the standard error will be a combination by standard statistical procedures of the standard errors for the individual set (see note h).

(e) *Random Counting Errors.* Include in this column, as one entry for a single source in each set of observations, the combination of errors arising from the statistical nature of counting; the error due to the background correction should specifically be included in the combination.

(f) *Random Errors Due to Weighing, Aliquoting and Others.* Include *random errors* of weighing, making aliquots and/or other operations associated with source preparation as distinct from errors which may be regarded as systematic.

(g) *Systematic Errors.* Include in this column an estimate of the *systematic error* associated with some or all of the following: weight or volume of source, weight or volume in dilution factors; adsorption (e.g., loss of activity to walls of containers); source drying (e.g., sublimation losses); slope of counting plateau (for 4π counting); corrections for dead time (τ_D), coincidence-resolving time (τ_R), self-absorption, source-mount absorption, half-life (see note j); implications of decay scheme and any others.

In practice a small number of these errors will be predominant and only those should be listed.

(h) *Mean Activity for Each Dilution.* State the mean activity for each set of observations used to give the result in column c.

(i) *Other Comments.* Include comments, if any, on the chemical stability of the solution. State typical value of ϵ_s for coincidence measurements.

Part 2, Corrections Applied

(j) *Decay.* State range of corrections applied. When half-life corrections are necessary, the half-life value used should be that stated by the distributing laboratory. It is suggested that, where possible, laboratories should make measurements before and after the reference time in order to reduce errors introduced by decay corrections.

If the measuring laboratory feels that the half-life quoted by the distributing laboratory is in serious error for a particular sample, a note on the measured half-life should be included here.

(k) *Background;* (l) *Dead-Time;* (m) *Accidental-Coincidences.* State range of corrections applied, identifying separate sets if necessary.

(p) *Other Corrections.* For example, corrections due to extrapolation in liquid scintillation β counting, density, internal conversion of γ rays, fluorescence yield. Please quote formula used in reducing coincidence data.

Part 3, Source Preparations

Please enter in the appropriate rows, details of the methods employed. If necessary, subdivide vertically for different methods.

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*These references are not cited in the report. They are concerned mainly with gas counting and 4π counting.

2. Low-level Radioactivity in Materials and its Relation to Radiological Measurements

2.1. Introduction

At the present time, increasing attention is being given to the measurement of very small quantities of radioactivity in human beings, foodstuffs, and miscellaneous organic and inorganic materials. These measurements may be concerned with, for example, (a) specific problems of radiation protection, involving the comparison of measured levels of radioactivity with the maximum values recommended by the ICRP, (b) longer term investigations, having as their aim the establishment of "normal" levels of radioactivity, against which any future changes can be judged, (c) applications of nuclear chemistry including the use of activation analysis, in the study of trace elements in human and animal tissues. Specific activities of interest will frequently be less than 1(dpm)/g.

It will be understood that the levels of activity studied in such measurements are very frequently much lower than the maximum permissible levels set out by the ICRP and widely adopted in legislation concerned with radiation protection.

In the design of measuring systems for the estimation of low levels of radioactivity, efforts are made to increase the efficiency of the detector and to reduce its background response. Relations between these parameters are considered in section 4.2. Radioactivity in the environment, or in the constitutent parts of a detector may be a major source of background and so, despite any improvement in the efficiency, may limit the precision of measurements of very weak samples, or of the radioactivity in human beings. Furthermore, radioactive contamination in reagent chemicals may lead to difficulties in preparing weak samples for assay.

In the past few years many data have been published on the *radioactivity of the biosphere* (see references). Far less detailed attention has been given to the systematic study of radioactivity in the inanimate environment but useful data have been published by a number of authors. Recently DeVoe (1961) prepared for the U.S. National Academy of Sciences a detailed report on contamination problems, using information supplied by many workers in the U.S.A. and Canada.

In the present report, a general, brief, description is given of some low-level radioactivity measurement techniques, and an attempt is made to review published work and data prepared by the ICRU, or submitted to them, on radioactive contamination in constructional materials, reagent chemicals, and related substances.

2.2. Low-level Measurement Techniques

Basic features of a number of important techniques at present in use have been summarized in table 2.1.

2.2.1. Alpha-Particle Measurements

For the measurement of very weak alpha-emitting samples, techniques such as those developed by Mayneord and his group (Mayneord, Radley, and Turner, 1959) have proved especially valuable. In their simple method, a layer of ashed or powdered material is spread on a thin layer of "nickel-killed" zinc cadmium sulphide, covering the bottom of a shallow tray, and scintillations are detected by a multiplier phototube. By careful choice of materials, the background counting rate for an area of nearly 100 cm^2 may be 2 alpha counts per hour. It has also been possible to determine the proportion of counts due to the thorium and uranium series by determining the number of closely spaced pairs that occur due to successive decay of thoron and ThA ($t_{1/2}$ThA=0.14 sec). More recently, these workers have developed large cylindrical pulse-ionization chambers for alpha spectrometry in which the alpha-emitting sample is sprayed in a thin layer on to an aluminized sheet of cellulose acetate forming the outer electrode (Mayneord and Hill, 1959).

2.2.2. Beta-ray Measurements

Counting methods used for assaying very weak beta-emitting samples have evolved from techniques developed some years ago by Libby and his associates for the measurement of natural C^{14}. The contribution from cosmic radiation to the background of the sample counter is largely eliminated by operating this counter in anti-coincidence with surrounding cosmic-ray detectors. The whole detecting system is enclosed in a massive shield made of materials as free as possible from radioactivity. Careful selection of materials used for the construction of the counter, its shield, and the sample mount has resulted in end-window (2.5-cm diam. or more) counters of high efficiency with backgrounds of less than 1 cpm (Grummit, Brown, Cruikshank, and Fowler, 1956; Bryant, Chamberlain, Morgan, and Spicer, 1957; Mayneord, Anderson, Bentley, Burton, Crookall, and Trott, 1958; Le Gallic, Grinberg, and Thern, 1960; Manov, 1960; Parker, 1960). Reliable commercial equipment of this sort is now generally available. Grinberg and Le Gallic (1961) have shown that an assembly of two gridded counters, with windows 10 cm in diameter is very useful in the measurement of low-level samples. Large-volume gas counters with backgrounds of a few counts per minute have been constructed by a number of workers, principally for C^{14}-dating purposes (Fergusson, 1955; deVries, 1957).

2.2.3. Gamma- and X-ray Measurements

For the assay of radioactivity by the detection of gamma or x rays, many thallium-activated sodium iodide, plastic- or liquid-scintillation sys-

TABLE 2.1. *Some techniques for measuring very weak radioactive samples and radioactivity in human beings*

Purpose of measurement	Detector	Form of sample	Method of identification	Typical shielding			Typical values for		Ref [3]
				Cosmic ray mesons	x & γ ray	Other	Background	Sensitivity [1,2]	
Assay of α-emitting samples: with limited analysis	ZnS(+mult. phototube)	Ashed or ground	For Th series by closely spaced pairs (see 2.2.1)	----	----	----	~3 cph	B.E.A.: 0.12 pc/g (Thick sample, ~11 cm diameter)	(a)
Assay of α-emitting samples: with identification	Pulse ionization chamber	Sprayed suspension on cellulose acetate	α-spectral analysis	----	----	----	6–10 cph per spectral line	B.E.A.: 0.1 pc in 1.5 g. sample, spread over 15,000 cm²	(b)
Assay of β-emitting samples	End-window G.M. or prop. counter (2-cm diam. window)	Solid precipitate	Prior chemical separation, confirmation by half-life or absorption curve	Anti-coincidence ring	10 cm Pb or 20 cm steel inner Hg ring	Borated wax neutron shield (esp. for gas counters)	~0.5 cpm	B.E.A.: 1.5 pc K^{40} in potassium salt (wt 60 mg, area 1 cm²)	(c)
Identification and assay of γ-emitting samples	NaI(Tl) crystal (e.g. 7.5-cm diam. x 7.5-cm high)	Solid or liquid. 2 liter samples in annulus	γ-spectral analysis	----	10 cm Pb or 20 cm steel (+0.75 mm Cd + 0.5 mm Cu)	----	~200 cpm above 100 kev (for 7.5 cm x 7.5 cm crystal)	"Minumim dectectable" ~4 pc Cs^{137} (in absence of larger activities) in 16h sample count +16h background count)	(d)
Assay of γ-emitting nuclides in human body: with identification	NaI(Tl) crystals e.g. (i)one 17.5-cm diam., x 8.75-cm thick (ii) four 10-cm diam. x 5-cm thick	----	γ-spectral analysis	----	(i) 20 cm Fe+3.2 mm Pb. (ii) 10 cm Pb +0.9 mm Cd	----	25 kev–1.575 Mev (i) 535 cpm (ii) 2035 cpm	"Minimum detectable" ~1 nc of γ emitter distributed throughout body, in ~30 min subject counting	(e) (see also table 2.2)
Assay of γ-emitting nuclides in human body: with limited identification	Organic liquid scintillator e.g., 530 liter	----	limited γ-spectral analysis	----	10 cm Pb	----	200–400 cps for measurements of 1–2 Mev γ rays	Efficiency 10–15 percent for K^{40} γ(1.5 Mev). Determine human K to ±5 percent in 100 sec subject counting	(f)
Assay of γ-emitting nuclides in human body: with limited identification	Plastic scintillator e.g., 3 units 50 cm x 25 cm x 15 cm	----	limited γ-spectral analysis	----	12.5 cm steel + 6-mm Pb lining	----	49.7–1.2 (range of values for cps per channel, low to high energies)	Efficiency 4 percent for K^{40} γ (1.5 Mev) (B.G. 25.9 cps). Determine human K to ±3.3 percent in 100-sec subject counting)	(g)

[1] B.E.A.: Background Equivalent Activity (see section 4.2).

[2] Data given are illustrative and based on work cited in the references. It is particularly difficult to express the minimum detectable limit for measurements of human beings, in view of the universal presence of potassium and fall-out Cs^{137} in all persons.

[3] References:

(a) Turner, Radley, and Mayneord, 1958.
(b) Hill, 1961.
(c) See references given in 2.2.2.
(d) Iredale and Humphreys, 1957.
(e) (i) Miller, May and Marinelli, 1959; (ii) Rundo and Speight, 1961.
(f) Anderson, 1956.
(g) Burch, Hughes, Iinuma, Overton, and Appleby, 1962.

tems have been developed. For the identification and assay of gamma emitters at low levels of activity, or low concentrations in bulk samples, the high detection efficiency of thallium-activated sodium iodide and its excellent energy resolution make it the detector of choice for such measurements. As it is often desirable to carry out measurements on gamma-emitting samples in bulk, an annular system is usually adopted in which the sample surrounds the detector. Cylindrical sodium iodide crystals 7.5 cm in diameter by 7.5-cm long have proved particularly suitable for work with samples of up to 2 liters in volume. Recently, large well-type crystals have been recommended for low-specific-activity measurements for samples up to 50 ml in volume (Perkins, 1961; Parr, 1962).

2.2.4. Discussion

It will be noted that the significance of particular forms and levels of radioactivity in the materials of the detectors, associated equipment, and shields will vary considerably for these different techniques; the main features of the problem, with some illustrative data, are set out in table 2.2. Furthermore, if chemical processing of the sample is necessary, radioactive contamination in reagents could lead to serious errors in an estimation.

TABLE 2.2. *Sources of background in low-level measurements systems*

(i) *General Survey*

Technique [1]	Chief sources of residual background	Remarks
α (total α counting by scintillation method)	α activity in scintillator, mounting, and multiplier phototube	No direct β-detection.
α (pulse ionization chamber)	α activity in cellulose acetate source-mounting electrode and central electrode. α activity in filling gas.	High resolution for particular α energy.
β (end-window counting)	Activity (α,β,γ,x) in walls of detector, and in sample mount. X,γ radiation from shield. Cosmic ray mesons passed by anticoincidence shield.	α,β,γ radiation can all produce effects.
γ (NaI(Tl), plastic and liquid scintillators)	Activity in crystal and multiplier phototube. X,γ radiation from shield, and atmosphere. Mesons, neutrons and secondary radiation due to cosmic rays. Mesons, neutrons ($n\gamma$ in shield, p recoils in gas) and secondary radiation due to cosmic rays.	α,β,γ radiation can all produce effects. Spectral analysis can reduce background due to low energy quanta.

(ii) *Illustrative Experimental Results*—(1) *GM Counters* Ref. (a)

Technique	Conditions	Counter No.	Background counting rate (1000 min counts) cpm [2]		Remarks
			Total	With anti-coincidence	
GM end-window sample counter surrounded by anti-coincidence cosmic-ray shield. Notes: (a) Sample counter: Metal-walled; window 2.5-cm diameter, of duralumin, 7-mg/cm² thick; counter diam. 2.5 cm; length 3.5 cm; components selected by manufacturers for low radioactive contamination. (b) Shield counters: Copper cathode in glass, efficiency of shield 97%.	Conventional Laboratory with 5-cm lead shielding on counter	(i)	5.89±0.17	1.01+0.09 −0.07	Means of 6 runs (each 1000 min).
		(ii)	6.42+0.38 −0.27	1.30+0.09 −0.09	
	With the same 5-cm lead shielding, but now beneath 1 m of chalk in rock below basement lab.	(i)	4.95+0.08 −0.12	0.54+0.07 −0.04	Means of 4 runs (each 1000 min).
		(ii)	5.63+0.20 −0.15	0.87+0.08 −0.10	
	Standard conditions, i.e., 5 cm of lead beneath 1 m of chalk	(iii)	—	0.57+0.06 −0.006	Means of 4 runs (each 1000 min).
	Beneath 1 m of chalk but no lead		6.64	1.47	Single run.
	Floor level in basement lab. with 5 cm of lead		—	0.96	Single run.
	Floor level in basement, no lead		13.2	7.36	Single run.

(ii) *Illustrative Experimental Results*—(2) *NaI(Tl) Scintillation Counter* Ref. (b) (c)

Technique [1]	Source	Background analysis cpm (25 kev to 1.575 Mev)	
		1 crystal [3], 17.5 cm × 8.75 cm (i) (Ref(b))	4 crystals [4], 10 cm × 5 cm each (ii) (Ref(c))
Sodium iodide scintillation counters	Crystals (K^{40})	30	760
	Shielding	110	—
	Phototube(s)	100	400
	Cosmic rays	155	400
	Radon in shield	–	~75
	Unidentified	140	400
	Total	535	2035

References (a) Bentley (1961).
(b) Miller, May, and Marinelli (1959).
(c) Rundo and Speight (1961).

[1] Assuming shielding etc. as given in table 2.1.
[2] Plus and minus indicate range of observed values.
[3] Low potassium content (2 ppm).
[4] Potassium content of crystals 65 ppm.

It must be stressed that, to reap the benefit of the precautions that may be taken to reduce radioactive contamination in detectors and shields, much care in the design and construction of electronic circuits is necessary, to achieve a stable background. Minor fluctuations which are of little account in conventional radioactivity measurements become of major concern when measurements of very weak samples are being carried out, perhaps over periods of many hours, or even days.

It is important that workers in this field should have the opportunity of intercomparing their techniques of measurement, and for this purpose standards of low specific activity are required. Such standards can then be used, without further dilution, in calibrating low-level-counting equipment. Results of an intercomparison of measurements of low-level $Sr^{90}+Y^{90}$ solution samples, distributed by the CEA–LMR, are summarized in table 2.3.

TABLE 2.3. *Low-activity* $Sr^{90}+Y^{90}$ *Comparison*

Details of Samples

1. Distributed: CEA–LMR June 1961.
2. Reference date: June 27, 1961.
3. Composition of solution:

$$50\ \mu g/g\ Sr(NO_3)_2 + 50\ \mu g/g\ Y(NO_3)_3 \text{ in } 0.1\ N\ HNO_3.$$

4. Samples distributed: 50 ml of solution in sealed ampoules.
5. Solution assayed at the CEA–LMR by 4π counting: the concentration was found to be 8.4 pc/g.

Participating Laboratories

CEA–LMR, IKO, NBS, RCH, Isotopes Inc., New Jersey, U.S.A., Department of National Health and Welfare, Ottawa, Canada, A.E.A. Research Group, Woolwich Outstation, London, U.K., and U.S.A.E.C. Health and Safety Laboratory.

Methods

Five laboratories made a chemical separation of Y^{90} for measurement. Three laboratories measured the radioactive concentration of the solution without chemical separation.

Counters calibrated with standards of higher radioactive concentration, and supplied by NBS, NPL, RCH, or AERE, and IKO were used in the measurements.

Results

Results from seven laboratories were in the range 8.05 to 8.7 pc/g; the eighth was 11.1 pc/g.

2.3. Contamination Problems and Data

In considering this subject, a distinction will be made between "radioisotopic" and "radiochemical" contamination (DeVoe, 1961).

By radioisotopic contamination will be meant radioactivity due to an isotope of a normal constituent element of the substance concerned; radiochemical contamination, on the other hand, will be contamination due to radioactive chemical impurities in the substance. This distinction is obviously important, since in principle radiochemical contamination can be reduced by straightforward methods, but it will seldom be feasible to attempt to separate the isotopes of a particular element, once contamination has appeared.

A further distinction can be drawn between contamination due to naturally occurring and artificially produced radioactive nuclides. This distinction is, however, of limited value. Certain radionuclides, notably H^3 and C^{14} come into both categories. Changes in levels of particular nuclides in the environment may arise not only from the artificially caused total increases in the activity of those nuclides, but also from changes in industrial processes. Thus the specific activity of C^{14} in atmospheric carbon has been reduced by the rise in the combustion of fossil fuels (Fergusson, 1958), but increased as a result of nuclear explosions. The contamination found in aluminum has been attributed to the use of raw scrap recovered from aluminum aircraft, contaminated with alpha-emitting luminous paint from instrument dials. While, therefore, as will appear from the data presented below, artificially produced radioactive materials have led to contamination problems, they represent only one among many causes of the changes that can occur in levels of radioactive contamination.

The widespread sources of radioactivity in materials are the members of the uranium and thorium series and K^{40}. There is a great variation in the concentration of these and other radionuclides in various materials. It is, however, frequently necessary to define maximum levels for particular application, e.g., in preparing legislation, in planning tracer experiments and in testing materials; this problem is considered in section 2.9.

A further interesting and important problem in contamination occurs in the artificial production of radionuclides. Frequently, because of the relative half-lives and reaction cross sections of the radionuclides concerned, small traces of chemical impurities can lead to significant and undesirable levels of contamination. In the radiological field, for example, such a problem has arisen in the detection of Ir^{192} in some samples of platinum-sheathed gold grains, this radionuclide having been produced by neutron irradiation of traces of iridium in the platinum sheaths. Thus, the purity required in materials to be irradiated for the production of particular radionuclides will usually need more precise definition than is necessary in normal analytical or metallurgical work; this is especially true if the materials are to be permanently implanted in human tissues, or otherwise administered to human beings.

2.4. Radioisotopic Contamination of Substances

The possible extent of radioisotopic contamination has been assessed by DeVoe (1961), who has examined current information on the production of radionuclides, particularly those having half-lives greater than 60 days, in relation to the elements which are not radioactive in their normal isotopic composition.

From tables 2.4 and 2.5 (both based on DeVoe's

TABLE 2.4. *Possible radioisotopic contamination of the elements (Z: 1–85) with isotopes having half-lives of 30 days or more. Based on a report by J. DeVoe (1961)*

Element	Nat. Act.	CR Act.	$t_{1/2}$<30 days	A	FP	CP	CK
1 H		+		+		o	o
He			–				
Li			–				
Be		+		+		o	
5 B			–				
C		+		+		o	o
N			–				
O			–				
F			–				
10Ne			–				
Na		+		+		o	
Mg			–				
Al				+		o	
Si						o	
15P		+	–				
S		+		+		o	
Cl		+		+		o	
A						o	
K	+					o	
20Ca				+		o	
Sc				+		o	((o))
Ti						o	
V				+		o	
Cr			–				
25Mn				+		o	
Fe				+		o	
Co				+		o	
Ni				+		o	
Cu			–				
30Zn				+		o	
Ga			–				
Ge				+		o	
As				+		o	
Se				+	+	o	
35Br			–				
Kr				+	+	o	o
Rb	+					o	
Sr				+	+	o	
Y				+	+	o	
40Zr				+	+	o	
Nb				+	+	o	
Mo						o	
Tc				+	+		
Ru				+	+	o	o

Element	Nat. Act.	CR Act.	$t_{1/2}$<30 days	A	FP	CP	CK
45Rh						o	
Pd					+	o	
Ag				+		o	
Cd				+	+	o	
In	+			+		o	
50Sn				+	+	o	
Sb				+	+	o	
Te	+			+	+	o	
I				+	+	o	((o))
Xe			–				
55Cs				+	+	o	(o)
Ba				+		o	
La	+					o	
Ce				+	+	o	
Pr			–				
60Nd							
Pm				+	+		
Sm	+			+	+	o	
Eu				+	+	o	
Gd				+		o	
65Tb				+		o	
Dy						c	
Ho						o	
Er			–				
Tm				+		o	
70Yb				+		o	
Lu	+					o	
Hf				+		o	
Ta				+		o	
W				+		o	
75Re	+					o	
Os						o	
Ir				+		o	
Pt			–				
Au				+		o	
80Hg				+		o	
Tl				+		o	
Pb	+			+		o	
Bi	+			+		o	

Key to Table 2.4

Element = Atomic number, symbol for element
Nat. Act. = Naturally occurring radioactive isotopes, $t_{1/2}$>10 years
CR Act. = Cosmic-ray produced radioisotopes. (See DeVoe 1961, appendix VI.)
$t_{1/2}$<30 days = All radioisotopes have $t_{1/2}$<30 days
A = Radioisotopes with $t_{1/2}\geq$30 days commercially available
F.P. = Fission product radioisotopes with $t_{1/2}\geq$30 days
C.P. = Radioisotopic contamination theoretically possible
C.K. = Radioisotopic contamination known
– = no such contamination

\+ = potential contamination
o = widespread contamination
(o) = occasional reports of contamination
((o)) = contamination reported, but of minor significance

Note: Raising the half-life limitation to >60 days (as used in the DeVoe report) only eliminates Yb from the list of elements in which radioisotopic contamination is theoretically possible (column 7). As noted in table 2.5, radioactive isotopes of V, Mo and Nd have been reported having half-lives exceeding 10^{15} years. Contamination by Nd may therefore also be "possible."

TABLE 2.5.* *Naturally occurring radionuclides of Z<83 based on report by DeVoe (1961) Appendix V-A*

Nuclide	Half-life years (approx.)	Radiation	Specific activity	Percentage abundance
H^3	12.4	β		variable
C^{14}	5.7×10^3	β	16 (βpm)/g C	
K^{40}	1.3×10^9	β,γ	31.8 (βps)/g K 3.4 (γps)/g K	0.0119
Rb^{87}	6.1×10^{10}	β	42.2 (βpm)/mg Rb	27.85
In^{115}	10^{14}	β	low	95.67
Te^{130}	$\sim10^{21}$	double β	?	34.49
La^{138}	7×10^{10}	β,γ	0.07 (βps)/g La 0.6 (γps)/g La	0.089
Sm^{147}	6.7×10^{11}	α	13.3 (αps)/g Sm	14.9
Lu^{176}	4.8×10^{10}	β,γ	?	2.59
Re^{187}	$<10^{11}$ or 4×10^{12}	β	?	62.93
Pb^{208}	20	β,γ	variable	variable

*Notes:
1. Other long-lived radionuclides of lead are reported.
2. V^{50}, Mo^{100}, and Nd^{144} are also reported in appendix V-A (DeVoe 1961) as possibly radioactive with $t_{1/2}>10^{15}$ years.
3. Data on percentage abundance are from Strominger, Hollander, and Seaborg (1958).
4. These data have not been critically reassessed for this report and are included to show the range of values encountered.
5. With the exception of Pb^{210} all the above radionuclides are singly occurring.

data) it will be noted that for nuclides with Z less than 82 (all known nuclides of Z greater than 83 are radioactive), 10 elements are radioactive in their normal isotopic composition (table 2.5) and 51 others (see table 2.4, column 7) have radioisotopes with half-lives greater than 30 days. The longer-lived isotopes of four of these elements (Ti, Mo, Rh, Dy) cannot presently give rise to contamination as they are neither fission products nor are they yet commercially available. (However, Rh^{102}, $t_{1/2}$=102 days, was produced in some of the weapons tests of 1958 (Libby, 1959).)

Thus, there are 47 elements with Z<82 which are susceptible to radioisotopic contamination and 10 others which may have their natural radioactivity augmented. To the present time, general radioisotopic contamination has occurred in the following instances:

(a) There have been significant changes in the concentration of tritium in territorial waters and in the atmosphere.

(b) The tritium content of stocks of heavy water and deuterium will be variable, and high as a result of both bomb and reactor contamination.

(c) Small increases have been reported in the specific activity of "live" carbon, and the concentration of C^{14} in atmospheric carbon dioxide has increased.

(d) There is a general and continuing contamination of krypton, extracted from the atmosphere, with Kr^{85}.

(e) Ruthenium compounds after extraction are generally contaminated with Ru^{103} and Ru^{106}.

There have been a few other reports of radioisotopic contamination, e.g., the detection of Cs^{137} in cesium iodide crystals, but there is no general evidence for such contamination apart from the effects given above.

2.4.1. Isotopic Concentrations of H^3 and C^{14}

Nuclear weapons testing has released large amounts of H^3 and C^{14} to the environment, augmenting the natural levels resulting from cosmic-ray irradiation of the atmosphere.

2.4.1.1. H^3

Considerable data on the tritium content of various natural waters and atmospheric gases have appeared in the literature of recent years. It should be noted that, in terms of the "tritium ratio" (TR),[3] for TR=1 the radioactive concentration is 0.13 (dpm)/mol of H_2O or H_2.

Before extensive thermonuclear testing, cosmic-ray production maintained tritium levels with TR values of 5 to 10 in continental rain and surface-drainage water, and about 1 in ocean rain and surface water. During, and for several months after, periods of thermonuclear testing in 1954–59, values of the TR for rain water ranged up to several thousand. Continued testing and dispersal of bomb tritium has resulted in a gradual rise in the tritium content of all surface waters. By 1960-61, continental and surface ocean waters of the northern hemisphere showed about 20 times the cosmic-ray tritium levels and sustained concentrations having TR values of 50-200 were general in normal domestic-water supplies.

Tritium was first observed in atmospheric hydrogen in 1948 at a concentration such that the TR equalled 4000 (Faltings and Harteck, 1950). Since then there has been a continuing rise to a concentration with a TR equal to 5×10^3 in 1960 (Bishop and Taylor, 1960). Bishop, Delafield, Eggleton, Peabody, and Taylor (1961) and Bainbridge, Suess, and Friedman (1961) also report significant amounts of tritium in atmospheric methane, a mean value for the northern hemisphere troposphere in 1958 giving a TR equal to 2.2×10^4. While these concentrations are much higher than those observed in natural waters, they are of very limited significance with respect to contamination problems since hydrogen and methane occur at very low concentrations in the atmosphere and are not normally extracted

Tritium concentrates with deuterium in isotopic-separation processes and, in consequence, all deuterium compounds will have a significant tritium content. For example, heavy water produced from natural water having a TR of 10 (a rarity nowadays) will have an activity due to tritium of 500 (dpm)/ml. Much of the D_2O inventory of the world has been irradiated in nuclear reactors and has had its tritium content greatly augmented by neutron-capture reactions. Reprocessing and redistribution of this heavy water is likely to contaminate any heavy water that has not been especially produced and carefully segregated.

2.4.1.2. C^{14}

The total quantity of C^{14} in the atmosphere, oceans and biological material has been estimated to be 56 to 81 metric tons, of which about 87 percent occurs in inorganic matter in the oceans and 1.7 percent in atmospheric carbon dioxide (Trotter, Zelle, and Hollister, 1958). Libby (1958) estimated that about 10^{28} C^{14} atoms or 0.23 metric tons had been introduced into the stratosphere by weapons testing. This represents an addition of only 0.3 to 0.4 percent to the total reservoir, but mixing of atmospheric carbon with the total reservoir proceeds very slowly. For the period March 1955 to March 1958, Broecker and Walton (1959) found an increase of 5 percent per year in the carbon dioxide in the troposphere of the northern hemisphere. This increase is reflected in measurements of the C^{14}/C^{12} ratio in plant materials. Broecker and Walton found the concentration of C^{14} in atmospheric carbon dioxide to be 14 percent higher in mid 1958 than in 1953.

The average specific activity of modern terrestrial carbon of biological origin was found by Anderson and Libby (1951) to be 15.3 ± 0.1 (dpm)/g of carbon[4]; later workers have found somewhat lower values, e.g., Fergusson (1955), 12.5 ± 0.2 (dpm)/g of carbon. Anderson and Libby's value corresponds to radioactive concentrations of approximately 8 (dpm)/l of CO_2 and CH_4 and 16 (dpm)/l of C_2H_2 at NTP. These values will have been raised by increases in the tropospheric C^{14} concentrations noted above, if the C^{14} in the gas sampled is drawn from the atmosphere.

2.4.2. Effects of H^3 and C^{14} Levels on Measurements

It has been indicated above that tritium concentrations with a TR of 50 to 200 could be expected generally in domestic-water supplies in 1960–61. Such levels become of significance in large gas counters. For example, if a 3-liter carbon-dating counter were filled at NTP with acetylene made from water with a TR of 50, the tritium disintegration rate would be 0.9 dpm,

[3] "Tritium ratio" is defined on page viii.

[4] 16.1 ± 0.5 for modern inorganic carbon; 15.3 ± 0.1 for modern biological carbon.

which would just be significant. Some water supplies will be fed more directly by fresh rainfall and hence have considerably higher tritium levels following a weapon-test period.

Difficulties could arise from the use of contemporary raw materials for the preparation of carbon-containing substances having various counting applications. The specific activity of contemporary organic carbon, 15(dpm)/g, corresponds to 14, 6 and 4(dpm)/g of polystyrene, polyvinyl chloride and PTFE respectively, and surface activities of about 0.05, 0.02, and 0.01(dpm)/cm^2. Ethanol used as quenching vapor at 1-cm pressure in a 1-liter counter, could contribute 0.21 dpm. In plastic and liquid scintillators, modern carbon would introduce between 5 and 15(dpm)/g of scintillator. These effects are small except in the case of scintillators, and are usually avoided since most plastic materials used in detectors are of coal or petroleum origin (see also section 2.6.1.1).

2.4.3. H^3 and C^{14} in Reagent Chemicals

There have been no reports of abnormal concentrations of H^3 or C^{14} in reagent chemicals in general use. A C^{14} concentration of 15(dpm)/g should be expected, but many chemicals will be free of C^{14}, being of coal or petroleum origin. Compounds containing exchangeable hydrogen will reflect the tritium concentrations of waters used in their manufacture and, if hygroscopic, could have the tritium concentration of current atmospheric moisture. Brown (1961) has observed the following tritium concentrations in several samples of reagent potassium hydroxide:

Date	H^3 Content (TR)
May 1955	31
March 1959	42
January 1961	27

2.4.4. Radioisotopic Contamination in Krypton

The most complete case of radioisotopic contamination is that of krypton. The only source of krypton, the atmosphere, has been contaminated with Kr^{85} released in reactor fuel processing and nuclear tests over the past 16 years. Delibrias, Perguis, and Labeyrie (1961) report the following radioactive concentrations of Kr^{85} in atmospheric krypton:

Date	Radioactive concentration, dpm of Kr^{85} per ml of Kr at NTP
1955	1.1
July 1958	4.1
March 1959	5.0
March 1960	4.0
June 1960	6.9
October 1960	8.1

Obviously this gas is ruled out of any low-level counting system. There is no alternative source of supply and only a very limited stock of lower activity material (DeVoe, 1961).

2.4.5. Radioisotopic Contamination in Ruthenium

Chaston (1960) and DeVoe (1961) report that ruthenium extracted during the last 10 years has been contaminated with fallout Ru^{106}. A change in mining practice is expected to avoid this in the future for some sources of supply, but one manufacturer in the United Kingdom (Chaston, 1960) has arranged to store newly refined material for six years before sale.

2.4.6. Radioisotopic Contamination in Cesium

Van Dilla (1960) reports the detection of Cs^{134} and Cs^{137} in three polished uncanned cesium iodide crystals, each 7 cm in diameter and 7-cm long.

2.5. Radiochemical Contamination in Materials

It should be noted that, using the most sensitive types of sample counting equipment now available, a very large number of materials will be found to contain traces of radioactivity, frequently due to radionuclides in the U or Th series. Data collected by DeVoe (1961) and others give many references to this problem, and some details are set out below.

It is immediately apparent, when representative data now available are tabulated (table 2.6) that, in general, little evidence has come to light that artificially produced radionuclides have contributed significantly to the contamination of the materials considered here. During periods of nuclear-weapons tests unprocessed raw materials became contaminated by radioactive fallout, and indeed any materials left exposed to the weather may be expected to become contaminated on the surface with both natural and artificially produced radionuclides. For example, cleaning of surfaces of steel sheets by sand or shot blasting (using sand or shot which is not significantly radioactive), or similar techniques, is necessary for applications in low-background counting. The particular concern in this report must be with contamination which cannot be removed by simple mechanical means, and is thus probably incorporated in the material considered.

2.5.1. Co^{60} Contamination in Steel

Before considering the contamination problem generally, the particular matter of Co^{60} contamination in steel will be considered in a little detail. Experience here illustrates the difficulties that can arise in setting proper levels for industrial tracer studies, and the lack of general agreement on the control that should be exercised over such studies.

Metcalf (1960) has summarized the work carried out by the British Iron and Steel Research Association, over a period of some eight years, in using Co^{60}, enclosed in pellets, for assessing the progress of lining wear in blast furnaces. The release of active pellets from the lining as it wears can be detected either by a counter set up outside the shell of the furnace or by detection of radioactivity in an iron sample taken during casting. It ap-

TABLE 2.6. *Typical data on radiochemical contamination*

Type of material	Due to natural radioactivity			Due to artificial radioactivity		
	Substance	Activity reported	Ref.	Substance	Activity reported	Ref.
Metals	Lead	Ra 3×10^{-3} pc/g	a	Mild steel	Co^{60} e.g.: 0.2–0.05 pc/g	a
	Mild steel	Th 1.6×10^{-3} pc/g	a		1 pc/g	i
	Stainless steel	Th (since 1953 due to use of rare earths).	b	Platinum	α (due to use as counting trays)	a,d
	Aluminum	Ra 0.3 pc/g	a	Lead	Fission products from scrap	a,c
	Beryllium	U	a			
	Brass	Ra (and Pb^{210} as isotopic contaminant) 0.3 pc/g brass.	a,d			
	Platinum	α emitters (due to use as counting trays).	a,d			
Reagents	Rare earths	Th, Ac	a	Sulphur	Zn^{65} 6000 (dpm)/g	a
	Phosphoric acid	Th, U	a			
	Organic reagents	U	a			
	Calcium chloride	Ra 1.7 pc/g	h			
	Calcium oxide	Ra	a			
	Hydrofluoric acid	U 10^{-6} g/g	a			
	Ammonium salts	U 0.5 (dph)/g	d			
	Hydrochloric acid	?β 1.7 (dpm)/l	e			
Miscellaneous	Paints	Ra 1 nc/l (Th, K much less)	d	Sodium iodide	Zr^{95}–Nb^{95} during nuclear weapon tests.	a
	Plastics	Ra (in those using metal powder as filler).	a	Activated charcoal	$?H^3$ 1500–5000 (dpm)/g of low energy β-emitter	d
	Sodium iodide	K^{40} 5 parts per million equivalent to 0.5 (cph)/g also Ra, Th	a,f etc.			
	Aluminum, titanium and magnesium oxide.	Ra, U, Th	a			
	Mica	K^{40}	a			
	Flux on welding rods	Th^{232}+daughters 20 pc/m rod Ra + daughters 1 nc/m rod.	d			
	Soft solder	Ra 30 pc/g	a,d			
	Multiplier phototubes	Th 31 pc Ra 370 pc K^{40} 730 pc } in a single tube	g			

References:
a. Information cited in Report by DeVoe, 1961.
b. Manov, 1959.
c. Sharp, 1961.
d. Peabody, 1961.
e. Brown, 1961.
f. Rundo, 1961.
g. Hughes, 1961; Iinuma and Burch, 1962.
h. Marlow, 1961.
i. McCall, 1961.

Notes:

1. These data and observations have been tabulated to illustrate the range of investigations at present being made; while there are many reports of the detection of the natural series of radionuclides in materials, fission products and other artificially produced radionuclides are reported very rarely except as superficial contamination.

2. Evidence on particular materials varies greatly e.g., tungsten is found contaminated with thorium by some workers, but satisfactory for low-level counting by others.

3. Estimates of the quantity of radium, thorium and artificial radionuclides likely to be disposed of as waste per annum in the United Kingdom have been made ("Control of Radioactive Wastes", Cmnd 884, 1959), (HMSO London, 1959). It is noted there, for example, that:

(a) Valve manufacturers use about 60 mc Ra per annum (1 μc $RaBr_2$ applied to cathode of certain valves).

(b) Possibly 90–300 mc Ra is disposed of as waste per annum from 37 establishments.

4. Problems of Radioactive Waste Disposal in the U.S.A., with special reference to reactor operations are considered in "Industrial Waste Disposal", 1959, U.S. Government Printing Office, Washington, D.C., 1959.

pears that this technique, while in common use in Europe for some years, has only been applied to a limited extent in the U.S.A.

Metcalf points out that limits must be placed on the quantities of long-lived radionuclides that may be dispersed in steel intended for general use. He discusses the necessary limits from the points of view of (a) exposure of human beings, (b) irradiation of film, (c) contamination in the constructional parts of radiation detectors and shields. Experiments showed that steel containing Co^{60} at a radioactive concentration of 40 μc/ton of steel, made up into cabinets, could reduce the life of x-ray film to 40 percent of that for unirradiated film. Again, Co^{60} at a radioactive concentration of 2 μc/ton of steel produced a 20-percent rise in the background of a Geiger-Müller counter made from this steel; Metcalf notes that for the low-level γ-scintillation systems much lower radioactive concentrations in the shielding walls are necessary.

Metcalf summarizes his recommendations thus: "It is recommended that the maximum permissible amount of Co^{60} in steel be set at 20 μc/ton, provided that a large proportion of the steel output can be guaranteed free from Co^{60} and that the steel for the sensitive parts of nucleonic instruments is taken from this portion or checked for radioactivity before use." He points out that practice has varied between countries, and refers to the conclusion of the U.S. Atomic Energy Commission, cited by Douglas (1954), that release of scrap steel containing less than 4.5 μc of radionuclides per ton would be "technically nonhazardous to sensitive industries".

Evidence for the contamination of steel intended for the shielding of low-background counting rooms has come from various workers; thus Salmon (1961) has reported Co^{60} activities which, if uniformly distributed, would give radioactive concentrations of 0.01 pc/g of steel. Hughes (1961)

reports detecting Co^{60} in a sheet-steel shield, although the steel had been certified as not coming from a furnace in which Co^{60} had been used. DeVoe (1961) reports measurements of Co^{60} in steel by several workers, who found values lying between 0.05 and 0.2 pc/g. McCall (1961) has found a sample with a radioactive concentration of 1 μc/ton.

Rasmussen, Bulkley, Harman, and Béique (1957) consider that the radioactive concentration of radium in a material to be used as a shield for low-background measurements of radioactivity in human beings should not exceed 0.01 pc/g, or equivalent, in γ-ray emission. It appears that this limit is quite frequently exceeded in present-day samples of steel, and this raises particularly serious problems in the construction of steel-shielded rooms requiring many tons of material. The testing of samples is time consuming and may be misleading. Consequently the practice of using for such rooms steel fabricated before 1945, e.g., from old battleships, appears to be necessary.

Practice with regard to the control of the use of tracers in industry varies from country to country, and it appears to be exceptional for such tracer applications to be governed by legislation, provided that no hazard to workers or the general population arises. Fortunately, it is also exceptional for such a long-lived tracer as Co^{60} to be used on any large scale, but it is evident that the scientific worker in the field of low-level radioactivity measurements must, at the present time, take his own precautions against such contamination affecting his work since it does not appear that adequate control is being exercised.

2.6. General Survey of Information on Radiochemical Contamination of Materials Used in Detectors and Their Shields

2.6.1. Detectors

2.6.1.1. Wall materials

Where "natural" levels only are concerned, the choice between materials such as brass, copper, mild steel and stainless steel, seems in the view of some workers, to be largely a matter of personal preference and, to a certain extent, luck. Some of the data collected on aluminum, copper, steel, and lead have been set out in table 2.7. Abson (1961) comments that all the commonly used metals contain radium, which can be detected most readily and conveniently by an alpha-activity measurement. Abson considers that mild and stainless steels are the best materials, and comments that they can be obtained with radium concentrations of about 0.03 pc/g, although occasionally pieces are encountered that have a much higher concentration. Abson reports much greater contamination in aluminium and brass, about 0.3 pc/g.

Since only a relatively small amount of material is involved in counter construction, this can be fairly easily checked by ashing or chemical separation. Alpha counting using pulse-height analysis

TABLE 2.7. *Some comparative data on levels of contamination in samples of metals*

Radiation	Technique	Ref.	Metal, with grade and measured activity								Units used to specify count rate
			Aluminum	Count rate	Copper	Count rate	Steel	Count rate	Lead	Count rate	
α	Ion-chamber counting over large internal surface area	a (8) 1933	Commercial	31	Machined Sand-papered Electroplated $CuSO_4$	9 8 13 to 160	Commercial	3	Solder (commercial)	2800	(cph)/100cm²
	Gas-flow prop. counter (internal samples)	a (51) 1956	?	22	?	.....	? Stainless	2.6 2.8	Soft solder (8nPb)	2600	
		a (1) 1960	(NBS 601) Other samples	13 to 16 16 to 79							
β	GM end-window counter (ashed sample)	a (29) 1956	"28" Superpure	9 4	Oxygen-free high conductivity (tubing)	0.05	.....	.....	Modern 16th cent. Solder 50/50 wire Solder 30/70 "Fluxite"	11 1 23 7	(cpm)/100 cm²
	GM end-window counter	b	.....	.....	.....	.....	.....	.....	Roman Britain S. Africa Missouri, U.S.A. Peru* Mexico English* English*	<2 8.4±0.8 2.5±0.7 29.4±0.6 11.0±0.6 6.1±0.6 12.9±0.6	
γ	5 cmx5 cm well type NaI (Tl)	a (38) 1960	"Alcoa" foil	0.5 ±0.5	.....	.....	.....	.....	.....	.....	(cpm)/g
	7.5 cmx7.5 cm NaI (Tl) 80 kev-1 Mev	a (44) 1959	.....	.....	Oxygen free Electrolytic foil	0.02 1.0	.....	.....	.....	.....	

References:
a. Report by DeVoe, 1961; ((8) 1933 etc., are references as cited by DeVoe).
b. Bentley, 1957.
*Electrolytically refined.
*Secondary lead.

is a particularly sensitive test, but it must be noted that detection of a single member of a natural series is not a reliable indicator of the total natural radioactivity, since manufacturing processes will have altered the equilibrium concentrations. For example, Grummitt and Roy (1956) found a radioactive concentration of radium of 0.9×10^{-3} pc/g in two grades of "Alcan" aluminum, but U^{238}/Ra^{226} α-activity ratios of 40 and 400 in contrast to the equilibrium value of 1. (The Ra^{224} and Ra^{228} content of these specimens corresponded to about 1 year's growth from 0.3 ± 0.08 parts per million Th^{232}, confirming the suggestion of selective radium removal during production.)

Oxygen-free copper, fused silica, and plastics are, at the present time, preferred materials for counter construction. Several commercially produced low-background counters use plastic for counter shell, window, and insulator. However, some adhesives and polyethylene sheet may introduce activity, and Brown and Grummitt (1961) report measurements on several adhesives showing beta activities ranging from 0.05 ± 0.04 to 12.5 ± 0.5(cpm)/g (measured as 2-g samples spread over 6.5 cm^2 and counted in approximately 20 percent geometry). The highest activities in materials from two manufacturers were found with the adhesives which were black (presumably carbon); some of these materials can add several counts per minute to the background of a counter. It is considered that this activity is due to radionuclides in the U and Th series that are in traces of minerals present in materials used for carbon-black production. It has been pointed out that whereas it might have been expected that the metal-filled adhesives would have shown the higher activity, the reverse was in fact found. It should be noted that traces of activity could be introduced into plastics from any contamination either in metal containers used in manufacture or in catalysts.

2.6.1.2. Solders and glass/metal seals

Glass used in Kovar glass/metal seals contains a few percent of potassium and hence some K^{40}. In addition, many graded seals contain uranium. These seals are often fixed to a counter by a soft solder which may contain radium concentrations of up to 30 pc/g. This additional background can be avoided either by suitable internal shielding of the insulator from the counting volume or by using quartz insulators fixed to the counter by epoxy-resin adhesives.

2.6.1.3. Wires

Samples of tungsten wire (0.025 and 0.012-mm diam.) (Crouch, 1961) have been reported to contain thorium equivalent to 1(cpm)/cm, but on the other hand Geiger-Müller and proportional counters have been made, using tungsten wire of 0.025 to 0.1mm in diameter, with total backgrounds of less than 0.1(cpm)/cm (Taylor and Peabody, 1961). Copper wires 0.12mm in diameter have been used in the grid of an alpha ionization chamber (Glover, 1961), and the background due to the wires could not have exceeded 0.0002(cpm)/cm.

2.6.1.4. Platinum and Aluminum

Similar data for these types of materials have been collected by DeVoe (1961), and from his report it is again evident that the natural-series radionuclides (U, Th), with K^{40}, represent the chief source of contaminating activity. DeVoe gives a short account of sources of relatively uncontaminated substances, noting in particular that virgin aluminum and platinum are found relatively inactive. However, DeVoe draws attention to the problem of alpha-emitting contamination in platinum; he points out that much platinum ware in which radioisotopes have been used is cleaned until it is free from radioactivity, as indicated by a standard laboratory survey meter, yet may still contain residual radioactivity outside the range of sensitivity of the instrument. If this platinum is scrapped, it will be added to virgin platinum and the resultant platinum may well be too active for use as a counting tray. Since alpha samples are frequently prepared for assay on platinum planchets, there appears to be a severe possibility of finding alpha-emitting contamination in platinum. At the present time no recommendations appear to have been made to guide users of platinum on disposal procedures, and problems of adequate checking are considerable. Certain manufacturers will carry out contamination checks on request, but this is not apparently a common practice as yet (Chaston, 1960). This service could be one task of the monitoring laboratories, the setting up of which is recommended in section 2.9.

DeVoe also discusses the source of activity in aluminum, noting that some has been found to be contaminated with radium and small amounts of uranium. He points out that since radium is immiscible with aluminum, it tends to form radioactive streaks or spots in the material. It is presumed that the contamination arises from scrap material, as discussed above (section 2.3).

2.6.1.5. Scintillation Detector Systems

Problems of H^3 and C^{14} in plastic and liquid scintillators have been discussed above (section 2.4).

In thallium-activated sodium iodide the chief contaminant is K^{40}, but crystals made in the USA may have a potassium content of only 1 part per million. Activity due to natural-series radionuclides has also been detected in thallium and adequate tests must be made of thallium to be used in such crystals. Data illustrating the contribution to the background of sodium-iodide-crystal systems due to radioactivity in the crystals and other causes have been given above (table 2.2). DeVoe reports that during atomic weapon tests in the USA sodium iodide was found to be contaminated with Zr^{95}-Nb^{95}. He also states that (1) there is some indication that Cs^{137} has been detected in sodium iodide, (2) radioisotopic contamination with fission product I^{129} ($t_{1/2}=10^7$ years)

has been reported, but is not expected to lead to problems, (3) cesium iodide has been reported to show occasional radioisotopic contamination with Cs^{137} (section 2.4.6).

Zinc sulphide is used extensively as an alpha-particle detector and there is evidence that a small amount of alpha activity is now present in commercially available zinc sulphide powder (Peabody, 1961).

Part of the background in scintillation detectors is due to radium and potassium in the glass envelope of the multiplier phototube and to potassium and uranium in the Kovar glass lead-in seals. Abson (1961) has quoted a typical radium concentration of 0.6 pc/g of Pyrex glass. The effects due to these contaminants can be reduced by using quartz for the main part of the envelope and by restricting any Kovar glass seals to the anode end of the tube. DeVoe reports extensive work in progress in the USA to develop a low-activity glass for use in multiplier phototubes.

The base and socket of multiplier phototubes can be a source of radioactivity. DeVoe reports measurements of a number of types of multiplier phototubes and of their components. Iinuma and Burch (1962) have given estimates of the activity in a particular 17.5-cm-diameter multiplier phototube of 31 pc Th, 370 pc Ra and 730 pc K^{40}.

2.6.2. Alpha-Particle Measurements on Materials

Table 2.8 gives data for natural activities determined with low-background alpha-detecting systems for a number of materials used in the construction of counters and shields and in associated work (Turner, 1961).

2.6.3. Shield Materials and Shield Construction

2.6.3.1. Steel and Lead

The problem of Co^{60} contamination in steel has been discussed above. It should be added that Co^{60} has also been found in lead; Salmon (1961) reports cobalt activities which if uniformly distributed through the material would be about 1 pc/g of lead and 0.01 pc/g of steel.

DeVoe points out that, like platinum, lead is often processed by addition of scrap lead to the virgin metal; if the scrap lead has been contaminated with fission products, the refined lead is likely to be radioactive. Sharp (1961) has referred to the sale of a large quantity of Eniwetok lead on the West coast of the USA as one source of contamination. Grummitt, Brown, Cruikshank, and Fowler (1956) and Manov (1960) have published data showing the differences in counting rate, for beta- and gamma-ray detectors, respectively, for various samples of lead. Manov's data show the importance of cleaning the surface, a reduction by a factor of ten being obtained by sandblasting both a steel and a lead brick. In Europe it is not too difficult to obtain sufficient quantities of "aged" lead, recovered from old buildings for shielding purposes; evidently if this lead had been adequately purified on first refining, so that only lead isotopes were retained, aging would drastically reduce the residual activity due to Pb^{210} (RaD $t_{1/2} \sim 20$ years). Thus at least two manufacturers in the United Kingdom keep a stock of lead from old buildings whose history is known and which had been in position for at least 50 years (Collins, 1961; Robson, 1961). On the other hand, for recently smelted metal, the activity depends on the origin of the ore. Bentley (1957) carried out measurements of the beta counting rate for various lead samples and found 1 to 8 pc/g for recently smelted lead, but 0.2 pc/g in a sample of aged lead.

2.6.3.2. Mercury

Kulp and Tryon (1952) used an inner mercury shield to surround a beta counter, within a shielded anticoincidence ring and this technique has been adopted by various workers. Sharp (1961) has concluded that triply distilled mercury is one of the few really clean shielding materials at present available; DeVoe does not refer to any studies of radioactivity in mercury.

2.6.3.3. Welding Rods and Electrodes

Miller and Steingraber (1957), Corcoran and Miller (1958) and Rundo and Emeleus (1961)

TABLE 2.8. *Radioactivity in materials determined by low-level α counting of ashed and powdered samples: Turner (1961).*

Item	Material	Radioactive concentration (α particles, in pc/g)	Identification
1	Sands	3 to 80	usually U+Th series Th/U: 2 to 3
2	Soils	7 to 36	U+Th series Th/U: 1 to 3
3	Chalks	0.7 to 1.0	U+Th series Th/U ~ 1
4	Analytic and reagent grade chemicals	0.5 to 2.0	Ra+Th series
5	Fertilisers	up to 300	usually U series only
6	Kaolin filler such as in Fireclays, Shoe whiteners, China crockery	up to 60	Th+U series
7	Polishing powders	up to 400	Th series only
8	Lighter flint	appr. 3000	Th series only
9	Incandescent mantles	appr. 100,000	Th series only
10	Tobacco ash	20 to 100	Ra+Th series
11	Bricks and cements	10 to 15	U+Th series
12	Plasters	10 to 110	sometimes U series only
13	Paper	up to 5	Th+U series
14	Oil paints	up to 90	Th+U series
15	Pencil lead	up to 15	Th+U series
16	Coal ash	up to 50	U+Th series
17	Ebonite, etc.	up to 10	
18	Woolen material	appr. 0.2	Th+U series
19	Rubbers	up to 15	Th+U series
20	Metallic lead	up to 30	
21	Detergents	up to 5	
22	Dental fillings	up to 350	U series only
23	Bauxite	up to 66	
24	Vitreosil	appr. 1.0	U+Th series
25	Disinfectant powders	up to 20	
26	Waxes	0.5 to 1.0	
27	Greases	up to 1.0	
28	Insect spray bases	up to 100	
29	Plastics	<0.1	
30	Zinc cadmium sulphide	<0.01	

Note: These data represent the ranges of activity encounted by Turner (1961) and are not based on comprehensive series of measurements of samples of each material.

have reported measurements of the radioactivity in welding rods and electrodes. No activity was detected in rods without flux, but flux was found to contain varying levels of K^{40}, radium and thorium; for example, concentrations of 2 pc of radium B+C, 0.4 pc of thorium and 0.02 g of potassium per gram of flux were reported by Corcoran and Miller (1959) for a heavy-construction-type electrode having a thick coating of flux. Evidently all welds in a low-background shield should be made using techniques such as gas welding, or carbon-dioxide-shielded welding, which require only bare wire.

2.6.3.4. Miscellaneous Materials

McCall (1961) reports that he has found radium or thorium contamination in all enamels and rubber based paints that he has measured. He also studied floor-covering materials based on asphalt, rubber, and vinyl plastic. He found that only pure vinyl floor covering was free of activity; the asphalt tile showed a strong radium spectrum. Peabody and Emeleus (1961) have made similar observations and have shown that several brands of paint (both lead undercoat and glossy top coat) all contained about 1 nc of radium 226 per liter and smaller activities of thorium and potassium.

2.7. Contamination in Reagent Chemicals

It is evident from the information studied during the preparation of this report that radiochemists working in the field of low-level counting have had relatively little difficulty with radioactive contamination of chemical reagents. It is customary for the radiochemist to rely upon blank analytical runs to reveal contamination in the reagent solutions in use, so that data on activity levels in specific chemicals are rather limited.

Bryant (1961), who has carried out much work on the separation and estimation of radioactivity due to fallout, reports that he has not found any contamination due to artificially produced radionuclides; activities found in some reagents were due to the naturally occurring series, and were found in lanthanum and yttrium compounds and in ammonium salts. Both alpha and beta activities were found troublesome in lanthanum used as carrier for lanthanides and actinides; the difficulty was overcome by substituting neodymium for lanthanum. Alpha and beta activities from the thorium series were identified in yttrium compounds, but were removed by iodate and sulphate separations from carrier solutions used in milking Y^{90} during low-activity determinations of Sr^{90}. Supplies of ammonium nitrate and acetate have been found by Bryant to contain uranium in very small amounts (about 0.5(dph)/g). Bryant remarks that the level of activity tolerated in carriers for this type of work on fallout is 50 (dpm)/g of element.

Marlow and Stockmann (1961) made a preliminary survey of a number of chemical reagents using a gamma-counting technique; similar studies have been made by Leddicotte (1960), Manov (1960), and Fix (1961), and details are given in the report by DeVoe (1961). Leddicotte, Marlow, and Stockmann, used well crystals of 5 cm×5 cm and 7.5 cm×7.5 cm (diameter×height) respectively.

Marlow and Stockmann found (for integral counts above 100 kev) more than 100 (cpm)/g for three samples of ceric ammonium nitrate and lanthanum nitrate and between 10 and 50(cpm)/g only for potassium salts. For the other samples studied (having weights generally between 2 and 10 g) the net counting rate due to the sample rarely exceeded twice the standard deviation of the measured counting rate. From their measurements on potassium salts, it appears that the efficiency of their system, for quanta of energy 1.5 Mev, is about 35 percent; samples giving a net counting rate of about 10 cpm (twice the standard deviation for most of the measurements reported) might therefore have an activity of about 15 pc in terms of gamma radiation of this energy.

Leddicotte identified the source of the activity found in some samples he studied. He found Lu^{176} in a sample of lutetium oxide, possibly identified Sc^{46} and Co^{60} in a sample of aluminum oxide and Sc^{46} in samples of oxalic acid; activity found in other samples is attributed to radionuclides of the U or Th series.

In DeVoe's report several tables are included illustrating the wide range of activity that can be detected in cerium salts; for example, Salutsky, Kirby, and Quill (1955) measured the alpha counts per hour from 2- to 5-mg mounted sources prepared from cerium salts, and found alpha activities ranging from 6 to 1245(cph)/mg of cerium.

2.8. Contamination Problems in the Manufacture and Storage of X-ray Film

The x-ray- and photographic-film industry can be seriously affected in several ways by environmental radioactive contamination. These include effects of (a) airborne contamination entering the processing works, (b) activity in the materials used in the film base and emulsion, (c) contamination in packing materials, (d) activity in steel storage cabinets (as discussed above).

Reports were submitted to the ICRU from two manufacturers in Great Britain, Messrs. Kodak and Ilford Ltd. The methods used at Kodak Ltd., have been published by Herz (1957), who describes the techniques for detecting fission products as well as natural radioactivity in filters on the air intake to the plant. These techniques were sufficiently sensitive to detect the fallout from the Nevada test explosions in 1952. Herz notes that the half-lives of the natural radioactive materials collected (RaB and RaC~20 min) and ThB (10.6 hr) are relatively short, whereas that of fission products is about 10 days. Consequently, failure to filter out fission-product activity could be more troublesome, and automatic devices have been installed permitting the identification of

fission products on the filters 5 hours after collection. Herz also described the technique used for detecting radioactivity in the cardboard or paper supply, pointing out that if such material is contaminated its use may be disastrous, particularly in long storage with photographic material of high sensitivity.

Lee (1961) of Ilford Ltd., reported various studies of radioactivity in packing materials. He stated that he had found no evidence of activity in the paper used in contact with the film, nor in the gelatin. However, spots were sometimes found, due either to activity in the film base, or packing materials. Particular problems had been experienced with Dutch strawboard, the difficulties having commenced in 1952. Subsequently several changes had been necessary in sources of supply of strawboard, or equivalent materials. It has been found that where two pulping processes were required in manufacture and accordingly plenty of water was used, activities were much lower. Chipboard, particularly low grades in which almost any waste material may be used to form the pulp was especially bad, presumably because there was a risk of incorporating contamination from recovered waste used in making the chipboard. Paper was almost free of activity because of the greater purity of the raw materials and perhaps the greater speed at which raw paper was run in production. Lee noted that the first complaints from users had arisen in 1958; at that time it had been thought safe to use strawboard for the boxes, but in one run of 600 boxes of film 339 spots were detected. On changing to wood pulp packing material only 10 spots were found in 637 boxes. Using chipboard they had detected 11 spots in 20 boxes. Routine tests of samples of film are made by the manufacturers; it was pointed out that the problem is most severe for x-ray film. The life of unexposed film is expected to be 1 year, but some users do store for longer periods.

2.9. General Comments

An attempt has been made in this part of this report to draw attention to the main fields of work where radioactive contamination, at low levels of activity, can lead to difficulty. Evidently, levels that can be tolerated will depend on the use to which a particular material is put. Problems that arise in making recommendations are illustrated by the examples given in table 2.9.

It will be noted, in particular, in table 2.9 that the lowest radioactive concentration is required in materials to be used as massive shielding for counting equipment. Furthermore, the level suggested by Rasmussen, Bulkley, Harman, and Béique (1957) is 1/2000 of that proposed as a limit (with various restrictions) for Co^{60} contamination of steel resulting from certain industrial tracer applications (Metcalf, 1960) (see 2.5.1). It is of great importance at the present time that low-level counting facilities, for human subjects and samples, should be widely available both for the purposes of long-term research (particularly related to the work of the ICRP) and for general monitoring of subjects and the environment. The addition of substantial quantities of such a long-lived tracer as Co^{60} to steel in production has already led to this important and convenient material being ruled out as a bulk shield by many workers. The problems of adequate testing and control are formidable, but with cooperation from the steel producer it should be possible to ensure that the relatively small quantities of steel required for this work can be obtained free from Co^{60} contamination.

It must be emphasized that in present legislation concerned with radiation protection, the levels of activity regarded as the minimum which need be considered far exceed those that can be tolerated in many materials used for low-background counting. Thus, in the U.K. Draft Factory Regulations (1960), a material containing less than 2000 pc/g is not regarded as radioactive yet this level is 2×10^5 times higher than the maximum level recommended for the radium content of shielding materials given in the recommendations of Rasmussen, Bulkley, Harman, and Béique (1957), and about 100 times greater than the level which Bryant has regarded as tolerable in carriers to be used in estimations of radioactivity in fallout, and similar work.

A number of useful recommendations were made by the U.S. National Research Council following the appearance of the report of DeVoe, which the Council had sponsored. These recommendations covered two main fields (a) the provision and development of adequate, standardized, monitoring facilities, set up and maintained by a particular laboratory; this laboratory would be responsible for testing a wide range of materials and chemical reagents, and for the study of sources of contamination; (b) the introduction of better

TABLE 2.9. *Some recommendations on maximum permissible radioactive concentrations as assessed for technical reasons, (i.e., for effects in scientific work, not from point of view of health hazards).*

Author	Topic	Proposed maximum radioactive concentration, pc/g
Rasmussen, Bulkley, Harman, and Béique (1957).	Maximum satisfactory level of contamination in a material for shielding an enclosure used in low-level measurements on humans.	0.01 (radium or equivalent γ emission)
Hours (1961)	Co^{60} in steel: proposal for maximum permissible level (for use in France).	2
Douglas (1959)	USAEC. Release of contaminated scrap steel to be "technically non-hazardous to sensitive industries".	4.5 (radionuclide)
Metcalf (1960)	Maximum permissible amount of Co^{60} in steel "provided that large proportion of the steel output can be guaranteed free from Co^{60} and that steel for sensitive parts of nucleonic instruments is taken from this portion or checked for radioactivity before use".	20
Bryant (see Peabody, 1961).	Radioactive concentration that can be tolerated in elements used in carriers for fallout estimations and similar work.	25

central control of "isotope" applications, so that practices which result in the introduction of radioactivity into industrial products are assessed, in order to determine whether possible long-term contamination difficulties can be justified by the expected benefits.

These are clearly valuable recommendations. In considering central control of "isotope" applications, it has been noted how tenuous this control can be, for example, in the Co^{60} work discussed above. Natural radioactivity can also pose serious problems, e.g., due to the activity of the flux in welding rods, or the general use of scrap that may be contaminated with discarded luminous-watch and instrument dials.

In France at the present time Le Laboratoire de Mesure des Radioéléments (Saclay) is attributing "quality labels" to various materials submitted to them. Seven classes are being utilized, designated by letters A–G, covering a range of activities from 0.002 pc/g to 196 pc/g; the ratio of the upper to lower limits in each class is 5:1. Thus materials are being classified from the point of view of their radioactive concentration, and it should be possible for prospective users of such classified materials to choose categories suitable for particular parts of shields, or radiation detectors by applying proper weighting factors.

The difficulties experienced in assembling material for this report, and the problems of comparing data obtained in various ways, lead us to the conclusion that there is a strong case for the setting up of a number of laboratories having major responsibilities on the lines proposed by the U.S. National Research Council. Such laboratories could provide at regular intervals reports on current contamination problems and could probably play a considerable role in assessing new proposals for large-scale tracer experiments.

2.10. Bibliography

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3. Availability of Radioactivity Standards—Present and Future Requirements

3.1. Present Availability

The sources presently available are presented in the following sections.

3.1.1. Beta and Gamma Emitters

The availability of radioactivity standards of the important nuclides emitting beta and gamma radiation is at present reasonably adequate, at least in the microcurie to millicurie range. In general, these are distributed as solutions, which is the most satisfactory form for most purposes. Sources containing known quantities of radium are also available from several laboratories.

Both H^3 and C^{14} present special problems because the chemical form of the material must be related to the counting method used. For C^{14} table 3.1 lists the calibrated material that is available. Future efforts in the calibration of C^{14} samples should be directed to increased accuracy rather than to increased diversity of material.

Normally measurements of H^3 are made either by gas counting or by scintillation counting, although in some cases counting of solid samples is found useful. The Radiochemical Centre, the National Bureau of Standards, and commercial firms in the United States offer calibrated samples of H_2O containing H^3; the Radiochemical Centre offers a calibrated H^3 sample which consists of hexadecane. Tritiated toluene is available as a calibrated material from the National Bureau of Standards and from a commercial source in the United States. This range of materials is satisfactory, but efforts should be directed towards improving the calibration accuracy.

TABLE 3.1. *Calibrated C^{14} samples*

Supplied by	Solid counting	Gas counting	Scintillation counting
U.S.A. Commercial firms	$BaCO_3$	$BaCO_3$ and Na_2CO_3 solution.	
	benzoic acid		benzoic acid
U.K. Radiochemical Centre.	polymethylmethacrylate.	polymethylmethacrylate	
	hexadecane-glycine	Na_2CO_3 solution	hexadecane glycine
France. Commissariat à l'Energie Atomique.			
U.S.A. National Bureau of Standards.		Na_2CO_3 solution	benzoic acid in toluene.

Note: The addresses of the governmental organizations are:
Radiochemical Centre, Amersham, Bucks.
Commissariat à l'Energie Atomique, Centre d'Etudes Nucléaires, Saclay.
The National Bureau of Standards, Washington, D.C. 20234

3.1.2. Alpha emitters

For radioactivity standards of alpha-emitting nuclides the situation is developing in a very satisfactory way. Calibrated deposited sources of Po^{210} are available from the National Bureau of Standards and from a number of commercial firms in Canada, the United States, and Western Germany. Solutions of polonium can be calibrated by various laboratories, such as the Radiochemical Centre, the National Physical Research Laboratory at Pretoria, and by the Institut für Radiumforschung in Vienna. However, some institutions consider Po^{210} to be objectionable for measurement as a solid, and its short half-life also constitutes a disadvantage. In the near future it is probable that the chief need for calibrated alpha-emitting material will be for the assay of plutonium in connection with nuclear technology and hygiene. Calibrated sources of Pu^{239} are available from the Radiochemical Centre[4] as solid deposits on metal discs, and as solutions. For many purposes these preparations will be excellent. A slight difficulty is met in the fact that the Pu^{239} is not, as a rule, isotopically pure. The Pu^{241} present produces Am^{241} by beta decay, which causes a slow but continuous increase in the alpha activity. For this reason it is desirable that the plutonium should contain only a very small admixture of Pu^{241} (with corresponding low concentration of Pu^{240}) and that the date of calibration and the relative content of Pu^{241} at that time should be indicated.

Some laboratories prefer, however, to calibrate Am^{241} instead of Pu^{239} and Po^{210}, as Am^{241} can be calibrated not only by means of quantitative alpha counting but also by coincidence counting. Over Po^{210} it has the advantage of not behaving as a volatile material. Calibrated sources of Am^{241} are available from the National Bureau of Standards. These are deposited sources, calibrated in terms of alpha particles emitted per unit time in a 2π geometry, rather than in disintegrations per second. Even so, a good approximation to the actual activity of the sample can be made. However, calibrated solutions of Am^{241} should also be made available.

3.1.3. Special Applications

Until recently, the production of radioactivity standards has been based mainly on the requirements for medical applications and for radiation hygiene. However, such standards will also be required for nuclear-energy developments. One serious need in this respect has already been taken care of by the distribution of calibrated samples of natural uranium and natural thorium by the New Brunswick Laboratory of the U.S. Atomic Energy Commission. For neutron-flux measurements the principal requirements are for calibrated samples of Co^{60}, Na^{24}, and Au^{198}, which fortunately are already available due to their important medical applications. For measurements of slow neutron fluxes, calibrated samples of Mn^{56} and of In^{116} would be most useful, the more so because the decay schemes of both of these radionuclides are quite complicated. (At present the decay schemes are too uncertain even to allow accurate determinations to be made by gamma measurement only.) However, the short half-lives of these radionuclides present serious problems. A distribution of In^{116} does not seem to be worth while in view of the difficulties involved. In the case of Mn^{56}, on the other hand, the excellent accuracy of the half-life (2.579 ± 0.005 hr) would permit a decay over a number of

[4] Presumably the accuracy of the calibration of plutonium sources could be much better than is offered at present. For most purposes, however, this improvement will hardly be desirable, in terms of the cost involved.

half-lives before measurement. Attention should be given to the question whether the material can be prepared with sufficient nuclear purity and it is desirable that experiments be made to this end. If purity permits, a distribution of Mn^{56} would be very useful, at least in countries where delivery is feasible. Alternatively, arrangement might be made for simultaneous measurements by the user and distributing laboratories. For radionuclides which are produced in detectors for fast neutrons, especially P^{32}, Na^{24}, and Co^{58}, suitable radioactivity standards are easily obtained. Calibrated samples of important fission products are also readily available.

A number of calibrated solid gamma-ray sources is being produced by the National Bureau of Standards. Even so, the availability of accurately calibrated sources of simple photon emitters, especially in the low-energy region, leaves much to be desired. It is recommended that a critical survey be made, and published, of the routinely available radioactivity standards.

3.2. Composition of Solutions

At present, most radioactivity standards are supplied as solutions.

It is recommended that the following information be included with all such solutions:

1. Activity per gram, accuracy and method of measurement.
2. Total activity or mass.
3. Composition of solution including pH, chemical form and concentration of carrier, chemical form of radioactive material, contamination by the presence of other radionuclides. (In the case of short-lived radionuclides, the information on this last point may have to be partially derived from previous experience with calibrated samples of the nuclide which have been similarly prepared.)
4. Methods by which the purity has been tested.[6] It may also be necessary to give some indication of the time for which a calibration is valid.
5. The decay-scheme data which have been used in the calibration should be stated.

In principle, the amount of radioactive impurities should be kept as low as possible and in any case low enough that the stated activity will be valid over a period sufficiently long for shipping and use. It is important that, in the case of short-lived radionuclides such as Na^{24} and K^{42}, care be taken in arranging the measurement and transportation time schedule. Laboratories calibrating and distributing these two radionuclides are requested to look very carefully into the shipping facilities and the effect of transportation time on the nuclear purity of the material upon arrival. (If Mn^{56} should be distributed, these points will be even more important.)

3.3. Required Accuracy

Comparisons of results of measurements on the same solutions of various radionuclides by many national and research laboratories have shown an overall spread in reported activity of about 5 percent (see tables 1.2 and 1.3). Where commercially available calibrated samples are concerned, it has not been generally demonstrated that a similar accuracy may be expected. Users of such materials should inquire from the suppliers how any accuracy quoted has been defined and achieved. It is essential that calibration procedures of laboratories distributing standardized material should frequently be tested by comparison with the national or, if necessary, with an international standardizing laboratory. For many applications in nuclear physics, medicine and radiation hygiene, an accuracy of a few percent is satisfactory. Occasionally a higher accuracy is necessary, e.g., when the sample is to be used to supply a known radiation field or known energy flux or in the absolute calibration of neutron sources (which itself attains to an accuracy of about 1 percent and therefore requires a somewhat better accuracy in the induced-activity measurement).

In order to increase the accuracy of calibration of the radionuclides by an order of magnitude, essential changes will have to be introduced into the techniques which are at present traditional, and standardizing laboratories should develop methods which promise an appreciable increase in the accuracy of the calibrations. In order to assess the accuracy of any calibration, it is necessary to use at least two methods. During the development of the calibration techniques for any radionuclide, the use of at least two completely independent methods is essential. For the routine preparation of accurately calibrated samples, the use of more than one method is desirable in order to check the accuracy of calibration.

Considerable discrepancies occur in the various descriptions of purity and in the use of terms such as precision and accuracy. The Commission intends, in cooperation with other bodies, to study these problems and to assist in the preparation of a code of practice.

3.4. Radioactivity Standards in Solid Form

The problem of calibrating solid sources of large dimensions was discussed in the 1959 Report of the ICRU (NBS Handbook 78, section 15.2, p. 73) and the opinion was expressed that calibration devices were at that time sufficient to cover existing needs. Very thin low-activity samples are, of course, easily prepared from solutions.

There is a need for accurately calibrated high-activity gamma-ray sources, probably in cylindrical form, but calibration is difficult due to self-absorption. These sources are required to provide gamma-radiation fields at different energies.

In discussing these sources a clear distinction should be made between calibration in terms of activity and calibration in terms of exposure-rate equivalent activity.[7] The activity is the disintegration rate in the source, which might be measured by dissolving the source and making a radio-

[6] The value of mass-spectrometer analyses has been neglected in this field and might well repay study.

[7] The use of the term "effective activity" for "exposure-rate equivalent activity" (or "equivalent activity" for short) is not encouraged.

active assay of the solution. The equivalent activity is equal to the activity of a point source of the same radionuclide which would give the same exposure rate at the same distance from the center of the source. Because of self-absorption and scattering there is no simple relation between the activity and the exposure rate at a fixed distance from a practical source.

Self-absorption factors cannot yet be calculated satisfactorily and experimental determinations are necessary. They are complicated by the fact that the activity distribution within a large-sized source may not be uniform.

The experimental determination of self-absorption factors should be associated with the measurement of specific gamma-ray constants. Work in this direction is going on at the Isotope Research Division of the United Kingdom Atomic Energy Authority.

3.5. Samples of Known Isotopic Composition

A few elements such as lithium, boron, and uranium, nowadays are occasionally, or even normally, sold having abnormal isotopic ratios. It is of the greatest importance that uranium of normal isotopic composition should be easily available for calibration purposes. The National Bureau of Standards at present provides U_3O_8 of normal isotopic composition.

There probably will be an interest in thorium samples with a known history, i.e., thorium samples which have been isolated from thorium minerals at a known date and possibly repurified at later dates.

3.6. Low-Level Measurements

3.6.1. Radioactivity Standards for Low-Level Measurement

Low-level measurements require the use of calibrated samples of much lower activity than the usual microcurie level. For many purposes such samples may be prepared by quantitative dilution, although these dilutions are not always easy to perform. In laboratories measuring low activity the handling of highly active material is undesirable. On the other hand, the preparation of very low-activity samples should not be done in an ordinary high-activity laboratory, because of the danger of cross contamination, and therefore requires a special laboratory.

Tl^{204} incorporated in a material which duplicates the sample in question is often used to calibrate counters for low-level measurements of fission-product beta activity. For this purpose it has suitable physical and chemical properties, but uncertainty as to its correct half-life may introduce errors if too long a time elapses between its calibration and use. It is suggested that the possible presence of impurities and the danger of adsorption losses be considered by laboratories which produce and calibrate this radionuclide. The possible use of Cl^{36} instead of Tl^{204} should be investigated, as it has approximately the same maximum beta-ray energy.

3.6.2. Samples for Testing Radiochemical Procedures

It is recommended that samples of various radionuclides which are important in fission-product control be distributed. These nuclides should be contained in the biologically important materials which are most frequently analyzed for contamination. The chemical state of the radionuclides in these samples should be as similar as possible to that of the normal radioactive contamination.

The chemical procedure to be used on the sample should be a close parallel to that normally used on the material under investigation. On the other hand, it would be most inconvenient to distribute dilute solutions like milk or urine. For this reason the following materials are suggested as most important ones for the radionuclides indicated:

milk powder (Sr^{90}, Sr^{89}, $Cs^{137}-Ba^{137}$, I^{131})
bone powder or bone ash (Sr^{90}, $Cs^{137}-Ba^{137}$)
dried urine or urine ash (Sr^{90}, Pu^{239}, $Cs^{137}-Ba^{137}$).

The samples provided should have an activity per unit weight appreciably higher than the material to be investigated, but not so much higher that dangers of contamination might arise, or that a different counting technique might be required. A sample with an activity per unit weight of about ten times the current average contamination level of the material in question would be suitable.

It would be useful if samples of this type could also contain those other radionuclides which in radiochemical analysis would be likely to contaminate the fractions isolated for measurement. In this respect a serious difficulty arises, both in the case of fallout and of possible reactor accidents, because the relative amounts of such contaminations will vary with time. The best known example is, of course, that of Sr^{89}, the activity of which becomes negligible compared to Sr^{90} about two years after nuclear fission has stopped. Nevertheless, it is recommended that attention be given to the possibility of providing samples containing such additional contaminating activities. An alternative approach would be to provide separate samples containing the additional radioactive contaminants but having the same general chemical composition. In the case of an alpha emitter, such as Pu^{239}, contamination by other radionuclides, which would be mainly beta emitters, could hardly present any complication and would probably not be worth adding.

It is understood that the International Atomic Energy Agency will make available various samples of environmental and biological materials such as soil, water, vegetation, animal bones and meat, and other animal products containing about ten times the average concentration of Sr^{90} and Cs^{137} found in these materials.

This decision of the IAEA is welcomed and it is suggested that samples of this kind be made available to a very large number of laboratories. No legal or administrative impediments to such a distribution are likely to be experienced as the quantity of radioactive material involved is extremely small.

4. Techniques for Measuring Radioactivity in Samples and Living Subjects

4.1. Introduction

This chapter deals with preferred methods of measurement of radionuclides for clinical and biological purposes. Most of the methods for which data are given are well established and are of satisfactory accuracy, particularly for sample counting. However, in the important field of *in-vivo* measurement the accuracy which must be accepted is lower and it is more difficult to recommend standardized procedures. Three aspects of *in-vivo* measurement are discussed: estimation of the radioactivity in the body as a whole, estimation of the radioactivity in an organ (such as the thyroid gland) and determination of the distribution of radioactivity within the body (scanning). These subjects are not treated in detail but, wherever possible, an attempt has been made to offer recommendations or guidance.

Useful sources of information on these matters include the recent "Manual of Radioactivity Procedures", NCRP–NBS Handbook 80 (1961), and several important symposia reports published by the International Atomic Energy Agency, Vienna. These and other general references cited in the text should be consulted by the reader for more detailed discussions of these subjects and for extensive bibliographies.

4.2. Sensitivity

In selecting the instruments most suitable for a particular measurement, many factors such as convenience and reliability enter into the choice, in addition to sensitivity. Various statistical methods of analysing data obtained with counting systems are available and may be applied in considering their relative merits. However, stability in the counting system is essential if such criteria are to be valid.

One valuable quantity is the *minimum combined time* (Loevinger and Berman, 1951). The total time which must be spent counting the sample and also the background in order to attain a specified precision will be a minimum if

$$\frac{T_{S+B}}{T_B}=\frac{\sqrt{S+B}}{B}.$$

Then, the minimum combined time, T $(=T_{S+B}+T_B)$, is given by

$$\sqrt{T}=\frac{1}{\epsilon}\left(\sqrt{\frac{S+B}{S}}+\sqrt{\frac{B}{S}}\right)$$

where S is the sample counting rate corrected for background, B is the background counting rate, and ϵ is the quotient of the standard deviation of S by S. It will be noted that the ratio of minimum combined times with two instruments will be a function of S and B, so that the better of two instruments for measuring weak samples may not be the better with strong samples. When S is small compared with B

$$\sqrt{T}=\frac{2\sqrt{B}}{\epsilon S}$$

and hence, since S is proportional to the counter sensitivity, f (the quotient of sample counting rate by activity), it follows that with very small values of S/B the shortest combined counting time is obtained with an instrument for which $\sqrt{B}/f$ is smallest.

The minimum combined time is somewhat complicated, and it is often useful to consider two simpler quantities: the *background equivalent activity* (BEA), and the *minimum detectable activity* (MDA).

The background equivalent activity, in the particular geometry used, is *that activity of a radionuclide which produces a reading of the instrument equal to its background reading.* (NCRP–NBS Handbook 80, section 2.5.4.)

$$\text{Background equivalent activity}=B/f.$$

Minimum detectable activity is defined as *that activity of a radionuclide concerned, which, in a given counting time, increases the reading of the instrument by an amount equal to three times the standard deviation of the background recorded in that time.* (NCRP–NBS Handbook 80, section 2.5.3.)

Table 4.1 gives values of background and background equivalent activity for typical detectors in four classes of measurement.

The minimum detectable activity is most easily computed for a counter in which the only fluctuations are those due to the random character of radioactive events. For that case

$$\text{Minimum detectable activity}=\frac{3}{f}\sqrt{\frac{B}{t}}=\frac{3B}{f\sqrt{N}}$$

where t is the time spent in counting the background and $N(=Bt)$ is the number of counts accumulated in determining B. It will be seen that, as deduced previously, the best counter at very low activities is that with the smallest value of $\sqrt{\frac{B}{f}}$.

The maximum measurable activities vary greatly with the apparatus, and are less clearly defined. For Geiger-Müller and scintillation counters the maximum measurable activity is in the range of 10^3 to 10^4 times the background equivalent activity, the upper limit being set by the acceptable dead-time correction. For large ionization chambers the maximum measurable activity may be about 10^6 times the background equivalent activity, the upper limit being set by the acceptable re-

combination loss. In practice the working range is usually limited by the associated current-measuring device (NCRP–NBS Handbook 80, footnote to table 7.1–1).

4.3. Preferred Methods of Measurement of Radionuclides

Techniques of measuring radioactivity in hospitals and biological laboratories are under continuous improvement with regard to sensitivity, accuracy, and convenience. This last is particularly important in studies involving large numbers of samples. Improvements are generally accompanied by increasing complexity of the apparatus, the subtleties of which can result in erroneous measurements if their operation is not fully understood. Table 4.1 and the accompanying notes present detailed information on the more important measurement techniques which may assist users to choose the methods most suitable for their purposes.

In table 4.1 the measurements are grouped into the following four classes:

I. High-level sources. Measurement of samples before administration to patients or animals.

II. *In-vivo* activity. Measurement of activity in the patient or animal (quantity or distribution).

III. Low-level samples. Measurement of samples or specimens from the patient or animal.

IV. Monitoring. Ancillary measurements concerned with laboratory monitoring and waste disposal.

About half the data quoted in table 4.1 has been taken from NCRP–NBS Handbook 80, tables 7.1–1, 7.1–2, 7.2–1, 7.6–1, and appendix B. The remainder has been assembled from various sources and then checked against current values in at least one radioisotope laboratory (Department of Radiology, Stanford Medical School). The data are intended to represent values which are reasonable at the present time, but not necessarily the best that can be accomplished with each technique.

TABLE 4.1. *Data on preferred methods of measurement of radionuclides for clinical and biological use*

Class	Purpose	Footnote	Detector	Background equivalent activity (μc)						Background B (epm)
				Beta-gamma		Pure beta			Bremsstrahlung	
				I^{131}	Co^{60}	H^3	C^{14}	P^{32}	P^{32}	
I. High-level sources	Calibration of bulk shipments & therapy quantities.	1	Calibrated well-type ion chamber.	1	0.2	–	–	0.04	100	(10^{-14} amp)
	Checking of aliquot for administration.	2	Simple, fixed-geometry GM counter.	4	1	–	–	–	200	40
		3	Simple, fixed-geometry scintillation counter.	0.1	0.2	–	–	–	10	200
	Dose-rate measurement on sealed sources.	4	Fixed-geometry ion chamber.	100	100	–	–	–	*	(leakage)
		5	Extrapolation ion chamber.	–	–	–	100	100	–	(noise)
II. *In-vivo* activity.	Uptake in tissue	6	Fixed-geometry scintillation counter.	0.2	0.3	–	–	–	20	900
	Whole-body counting	7	One or more liquid or solid scintillation counters.	*	–	–	–	–	*	*
	In-vivo distribution	8	Scintillation counter and mechanical scan.	0.04	–	–	–	–	–	60
		9	Two scintillation counters in coincidence.	–	–	–	–	–	–	*
	Relative surface activity	10	Small, hand-held scintillation counter.	0.01	0.02	–	–	–	0.6	100
		11	Small, end-window GM counter.	*	*	–	–	*	–	*
	Relative activity within tissue.	12	Needle-probe GM counter.	10	*	–	–	3×10^{-4} $\frac{\mu c}{ml}$	–	12
		13	Needle-probe scintillation counter.	*	*	–	–	*	–	*
III. Low-level samples.	Unprocessed excreta and other large samples.	14	Scintillation counter	0.07	0.05	–	–	–	30	1400
		15	Ring of large GM tubes	0.1	0.1	–	–	–	20	1000
		16	Well-type ion chamber	3	1	–	–	–	300	(10^{-13} amp)
	Blood and other small samples.	17	Well-type γ scintillation counter.	2×10^{-4}	2×10^{-4}	–	–	–	9×10^{-4}	200
		18	Well-type β scintillation counter.	–	–	–	–	4×10^{-4}	–	10
		19	Liquid scintillator	–	–	1×10^{-3}	4×10^{-3}	–	–	100, 40
		20	Thin-wall liquid GM counter.	5×10^{-4}	8×10^{-4}	–	–	4×10^{-4}	–	10
	Dry Samples	21	End-window GM counter.	5×10^{-4}	5×10^{-4}	–	8×10^{-4}	5×10^{-4}	–	20
		22	Gas-flow GM counter	1×10^{-4}	1×10^{-4}	1×10^{-4}	1×10^{-4}	1×10^{-4}	–	10
	Chromatographic and autoradiographic samples.	23	End-window GM counter	5×10^{-4}	5×10^{-4}	–	8×10^{-4}	5×10^{-4}	–	20
		24	Gas-flow GM counter	1×10^{-4}	1×10^{-4}	*	1×10^{-4}	1×10^{-4}	–	10
IV. Monitoring	Routine laboratory monitoring.	25	End-window GM counter	1×10^{-4}	1×10^{-4}	–	2×10^{-4}	1×10^{-4}	–	40
		26	Scintillation counter	0.02	0.03	–	–	–	2	700
	Disposal	27	Simple, fixed-geometry GM counter.	4	1	–	–	–	200	40
		28	Simple, fixed-geometry scintillation counter.	0.1	0.2	–	–	–	10	200
		29	Calibrated ion chamber	3	1	–	–	–	300	(10^{-13} amp)

–=not applicable.
*=no suitable data available.

Notes on Table 4.1

1. The data are for one design of well-type ionization chamber which has been developed by the National Physical Laboratory, Teddington, England, and is commercially available (Dale, Perry, Pulfer, 1961; Dale, 1961). These chambers are manufactured to close tolerances and are all of equal sensitivity to within 1 percent for γ rays and 3 percent for β rays. Calibration factors are available for a number of radionuclides. Activities up to several hundred millicuries can be measured, and with conventional electrometers the minimum detectable activity corresponds to approximately 10^{-14} amp; hence the minimum detectable activity is comparable with the background equivalent activity. Another well-type chamber for this purpose is commercially available in France (Engleman, 1960).

2 and 3. Any reproducible system is satisfactory, provided source-detector distance is not so short that positioning errors are important. If previous measurements are made by the supplier, the accuracy required is not high as the purpose is to check for order-of-magnitude errors and sample identity. (A well-type ion chamber (notes 1 and 16) can also conveniently be used for this purpose.) The quoted sensitivities refer to distances of 20 cm.

3. The data apply to a scintillation crystal, 2.5x2.5 cm, used with discriminator at 50 kev, distance 20 cm.

4. The data apply to an *R*-meter ionization chamber which gives exposure rate directly, but the chamber-source distance should not usually be less than 5 times the largest ion-chamber dimension. The well-type ion chambers (notes 1 and 16) can be very conveniently used for relative measurement of gamma-emitting sources of the same radionuclides, or of different radionuclides if the chambers are effectively "air equivalent" or are properly calibrated.

5. The extrapolation chamber is an absolute instrument, in the sense that exposure rate is determined directly in terms of geometrical and current measurements only. It is a specialized instrument, and its use requires special care and experience.

6. The sensitivities refer to a 5 x 5-cm crystal, at 25 cm with discriminator at 50 kev. A larger-diameter crystal reduces the minimum detectable activity, but does not appreciably change the background equivalent activity. A pulse-height analyser allows rejection of scattered and background radiation, and improves the sensitivity, no matter how this is specified. Either a scaler or a precision count-rate meter can be used for straight uptake measurements. For measurement of rapidly changing activity (e.g., cardiac output) a count-rate meter with a linear or logarithmic recorder may be used. It is necessary that both instruments have a fast time response, in order not to distort the response curve.

7. A variety of scintillation systems is used. See section 4.6.

8. The sensitivity for I^{131} refers to a 5 x 5-cm crystal with a 19-hole focussing collimator, and a 100-kev window centered at 364 kev; however window widths for scanning with I^{131} may be as small as 40 kev.

9. Measurement of the coincidences of the photons of β^+ annihilation requires two scintillation crystals and suitable coincidence circuitry. See section 4.5.

10. The sensitivity refers to a 1.25 x 1.25-cm crystal at 2 cm without discrimination. The relative γ-ray activity of two superficial regions (e.g., palpable thyroid nodules) is best examined with a small scintillation crystal with minimal shielding placed directly in contact with the areas. A simple power supply and either scaler or count-rate meter is adequate.

11. The relative β-particle surface activity of two regions (e.g., the anterior surfaces of the eyes) is best measured with a small end-window GM counter. No shielding is necessary. A simple scaler or count-rate meter is adequate The absorption of β particles is so great by even thin layers of over-lying tissue that it is difficult to give meaningful figures for the background equivalent activity.

12 and 13. The relative *in-vivo* β-particle activity inside a tissue (e.g., brain) is best measured with a fine probe. Both GM and scintillation probes are commercially available. A simple power supply and either scaler or rate meter is adequate.

12. The figure for I^{131} is for a distance of 10 cm, while that for P^{32} is in μc/ml.

14. The sensitivities refer to a 4.4 x 5.0-cm crystal, without a pulse-height analyzer, and a one-liter sample placed directly on top of the crystal. With this technique the reproducibility of the geometry, including the sample volume, is very important. A discriminator is useful to eliminate low-energy background counts, but it should not be used to eliminate radiation scattered in the sample or detector. A discriminator setting of about 50 kev is usually satisfactory.

15. This technique has the advantage of being simple, stable, inexpensive, and insensitive to variations in source position. The GM tubes can be connected in parallel.

16. The sensitivities refer to a well-type ion chamber constructed to accept a sample volume up to 4 liters (1 gal) at a time. While less sensitive than the preceding method, it is simple, stable, and insensitive to source position. It has the advantage of a very wide range of response, being suitable for sources up to several hundred millicuries, With conventional electrometers the minimum detectable activity corresponds to approximately 10^{-11} amp, hence the MDA $\approx$ BEA/10.

17. The sensitivities refer to a 4.4 x 5.0-cm crystal, containing a well 1.9 cm in diameter and 3.8-cm deep, with discriminator set just above phototube noise. A discriminator is adequate for single radionuclide counting. The discriminator should be set well below the photopeak, e.g., 200 kev for I^{131} and Cr^{51}, and 800 kev for Co^{60} and Fe^{59} are reasonable settings. For bremsstrahlung counting, the discriminator should be set just above the phototube noise. For multiple-tracer measurements, a pulse-height analyzer is almost essential.

18. The data apply to a 5-ml sample in a plastic test tube. Due to absorption of the β particles the results are very sensitive to sample volume and container-wall thickness. The advantage of an organic β-sensitive phosphor is its low γ-ray sensitivity and, as a result, its very low background. It is essential to eliminate low-energy background pulses with a discriminator. Since no peaks are found, it is very difficult to calibrate this detector in terms of energy.

19. This is a very sensitive method of counting, especially useful for the very low-energy β particles of tritium. The method requires elaborate counting apparatus and refrigeration. The liquid phosphor is easily poisoned (quenched) by an incompatible sample, so careful use of control samples, with and without activity, is necessary. The higher background applies to the counting conditions for tritium.

20. The data refer to a counter with glass walls 30-mg/cm^2 thick. This detector is usually a thin, jacketed, glass-wall tube for active liquids. These are stable and inexpensive GM tubes and are particularly suitable for beta particles of higher energy. They are, however, also very useful for lower energy $\beta\gamma$ emitters such as I^{131} and even for Co^{60} where the γ rays alone are recorded. Thin-wall metal GM tubes are also available for "wrap-around" sources.

21. The sensitivities refer to a counter with a 1.4-mg/cm^2 window. This detector is simple, stable, inexpensive and is outstanding for those techniques for which it is suitable. Halogen-filled tubes offer indefinite life, and a simple scaler and power supply are adequate. Reproducible geometry, including mass of sample, is essential.

22. The sensitivities refer to a windowless counter. This method is stable and reproducible and has the advantage of high geometrical efficiency, as a result of placing the sample inside the counter. Contamination of the detector is sometimes a problem. Special gases are usually used, but many counters will operate satisfactorily at higher voltages with natural gas (i.e., illuminating gas).

23. Comments on the detector in note 21 apply here. Chromatograms and autoradiograms can be examined manually, using a large detector with an aperture, or a small detector. Automatic paper-chromatogram scanning is provided by commercial equipment which moves an active strip past the detector slit synchronously with the chart

motion on a linear recorder, the latter receiving the output of a count-rate meter. The linkage between the active strip and the chart must be very reliable, or interpretation will be difficult. It is desirable that the recorder should periodically indicate fixed points on the chromatogram strip.

24. Comments on the detector in note 22 on the automatic scanner in note 23 apply here. For very low-energy β particles, especially tritium, it is necessary to have the active paper inside the counting gas. A convenient form of automatic gas-flow chromatogram scanner moves the paper through a narrow gap between two opposed counters, thus counting both sides of the paper.

25 and 26. A very simple scaler or count-rate meter is adequate for laboratory monitoring, though it is very desirable that it have an audible signal. It is generally most convenient if this equipment is a portable survey meter. For monitoring laboratory benches and glassware for β-particle contamination, the end-window GM tube with a window of about 1.5 mg/cm^2 is best for all β-particle emitters except tritium. For monitoring γ rays alone, a GM counter may be used, but a scintillation crystal is much more sensitive.

25. The data apply to the same detector as in notes 21 and 23, but for laboratory monitoring the detector is assumed to be lightly shielded, so the background is taken to be twice as large.

26. The data apply to the same detector as in note 3 but the discriminator is set just above the phototube noise, the distance to the source is 2 cm and the detector is assumed to be lightly shielded so that the background is taken to be four times as large.

27 to 29. For waste-disposal monitoring, it is only necessary to make an estimate of the activity. For this purpose a relatively insensitive, fixed-geometry detector is suitable.

27. The data apply to the same detector as in note 2.

28. The data apply to the same detector as in note 3, but the discriminator is set just above the phototube noise.

29. The detector is the same as in note 16 and is larger than the detector described in note 1.

4.4. Thyroid I^{131} Uptake Procedure

The measurement of radioactivity within the body of a patient or animal, class II in table 4.1, is much more difficult than the measurement of the discrete samples encountered in classes I, III, and IV. In general lower accuracy must be tolerated and it is more difficult to recommend detailed procedures that might be acceptable to everyone. However, in measurement of thyroid uptake which is a very widely practiced medical procedure, some degree of standardization of procedures would be highly desirable in order that uptake values may be compared from laboratory to laboratory.

In recent years, several groups have considered the various factors which determine the accuracy of an uptake measurement and some have made recommendations concerning procedure (Brucer, 1960; NCRP–NBS Handbook 80, 1961; IAEA, 1962). The most recent recommendations are those of a group convened by the International Atomic Energy Agency which met in Vienna in November 1960 and considered primarily uptake measurements with I^{131} carried out 24 hours after the administration of the dose (IAEA, 1962). The group recommended a method of measurement which would ensure that the results obtained would be comparable with those reached at other institutions using the same method, yet still provide adequate accuracy, even under rather primitive conditions. These recommendations are endorsed because it may be preferable to achieve agreement among laboratories than to strive for the highest accuracy.

The recommendations of the IAEA group, which deal with the physical aspects of the procedure, may be summarized as follows:

(a) In order to minimize the quantity of I^{131} to be administered to the patient, a scintillation counter with a crystal size of not less than 2.5 x 2.5 cm should be the instrument normally used.

(b) The visual field at the working distance (20–30 cm) should be preferably 12 cm and certainly not greater than 15 cm in diameter (fig. 4.1.).

(c) Collimation and shielding should be such that, if a point source of I^{131} is moved at the working distance, the counting rate falls to 50 percent or less as the distance from the axis increases by 20 percent, and to 5 percent or less as this distance increases by a further 20 percent. With a further movement of the source away from the axis, the counting rate should fall rapidly and remain below 1 percent of the maximum value (fig. 4.1.). Figure 4.2 shows the upper limits of counting rate outside the field as a fraction of that in the field.

(d) The contribution of scattered radiation from the patient to the total counting rate should be reduced to a minimum. This could be done either by placing in front of the detector a lead filter of 1.5-mm thickness or by setting the scaler at a suitable bias level, such as 280 kev. Scaler thresholds should be carefully checked.

(e) The volume of the solution to be used as the "standard" should be similar to that of thyroid glands frequently measured, e.g., a polyethylene bottle approximately 30 mm in diameter and filled with 30 ml of solution will be satisfactory. This bottle should be placed in a cylindrical neck phantom of 15 cm in diameter and of 15 cm in height, made preferably of lucite or perspex. The recommended position within the phantom of the hole for the bottle is illustrated in figure 4.3.

For the detailed version of these and other recommendations of the IAEA group, as well as for some comments intended to show the reasons underlying some of the points made, the original text of the recommendations should be consulted.

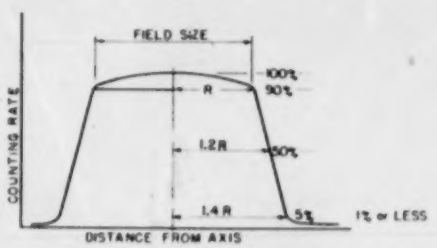

FIGURE 4.1. *Relative counting rate of detector vs. distance between the axis of the collimator and a point source at the working distance (see fig. 4.2) for an adequately designed collimator.*

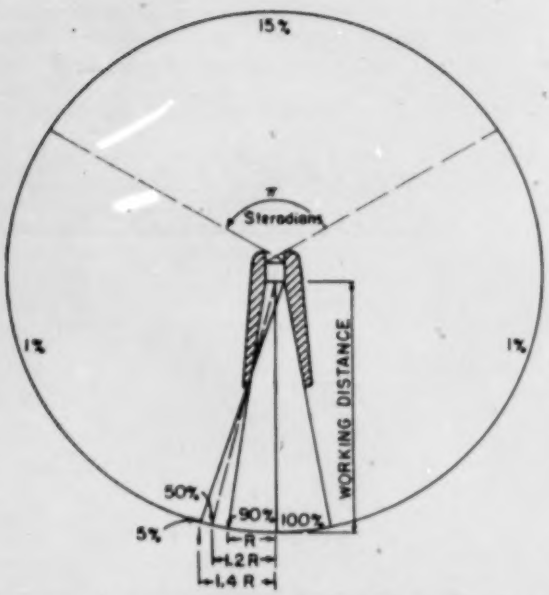

FIGURE 4.2. *Schematic drawing of crystal and collimator and of upper limits of counting rates outside the field of vision.*

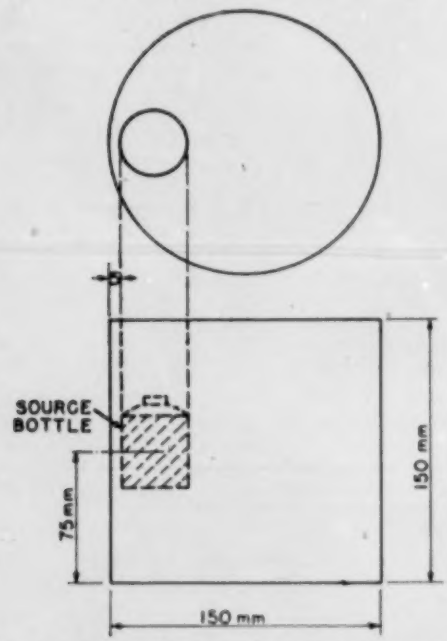

FIGURE 4.3. *Schematic views of the neck phantom and source bottle.*

4.5. *In-Vivo* Distribution Studies (Scanning)

4.5.1. Introduction

The determination of distribution of radioactive material *in vivo* by automatic scanning is a rapidly growing field and it is as yet too early to recommend standardized procedures. However, it is possible to consider some aspects of scanning, including some of the features that good scanning equipment should possess.

Scanning is performed with gamma-ray-emitting or bremsstrahlung-producing radionuclides and conventional circuitry, or with positron-emitting radionuclides and coincidence circuitry. These various techniques of scanning are reviewed in NCRP–NBS Handbook 80 (1961), and in IAEA–WHO "Medical Radioisotope Scanning" (1959). The present discussion is limited to automatic gamma-ray area scanning with a moving detector.

4.5.2. Detector

Gamma-ray emitters of low energy are commonly employed in scanning and for these sodium iodide crystals are the detectors of choice. The diameter of the crystal is limited by practical considerations of shielding weight, and the thickness is limited by the consideration that there is little gain in sensitivity beyond about 5-cm thickness (for I^{131}), while the background increases in proportion to thickness. Crystals 5 x 5 cm are a good compromise, although both larger and smaller crystals have been used successfully.

4.5.3. Collimators and Shielding

The design of the collimator controls the region seen by the detector. Since collimators generally provide improved resolution at the expense of sensitivity, at least three interchangeable collimators covering a range of resolutions and sensitivities will be found useful. For example, Dewey and Sinclair (1961) have shown that, under certain well defined conditions, a collimator having a very high resolution (e.g., a 91-hole focusing collimator) offers the best chance of detecting radiation from a region of less than 0.5 cm in diameter (and high activity is necessary); for regions between 0.5 cm and 1.5 cm in diameter a lower-resolution and higher-sensitivity collimator (e.g., a 19-hole focusing collimator) is best; for regions between 1.5 cm and 3.0 cm in diameter a more modest resolution and higher sensitivity collimater (e.g., a 7-hole focusing collimator) is best; and above 3.0-cm diameter the lowest-resolution and highest-sensitivity collimator (a single-bore collimator) provides the best chance of detection. The properties of a focusing collimator are that it has a sharp resolution and high sensitivity for small sources at the focal distance; therefore the distance between the collimator and the region to be detected is critical. These properties, however, lose their advantage in practice when the source is large. A slit collimator has advantages for profile scanning. Definitions and data on the resolution of collimators are given in NCRP–NBS Handbook 80 (1961); Brownell (1958); Dewey and Sinclair (1961); and Beck (1961).

In collimators of small angle of acceptance it is of the utmost importance that shielding of the remainder of the field should be of ample thickness; otherwise radiation penetrating the shield may destroy the resolving power. Some commercially available collimators are deficient in this respect.

4.5.4. Mechanical Features

A variety of mechanical means of moving the detector relative to the subject has been devised. The most widely used type of area scanner employs a detector which is moved over a region of the subject in a series of equally spaced parallel lines.

A versatile scanner should have the following mechanical characteristics:

(1) variable-scanning area up to about 40 x 70 cm;

(2) facilities for interchanging collimators;

(3) means of identifying the detector position with respect to the patient;

(4) variable line spacing;

(5) variable speed covering at least the range 0.1 to 1 cm/sec. Control over scan speed and line spacing makes the scanner useful for large areas, where with higher speed and wider spacing it should be possible to complete a scan on a patient in no more than 30 minutes. The line spacing chosen also depends on the resolution of the collimators used. Experience has shown that the speeds indicated cover the most practical range. However, the influence of scan speed on resolution and detectability has not been thoroughly studied. In certain situations, it might be found useful to scan the region of interest in a second plane at right angles.

4.5.5. Electronic Features

The detector is followed by the usual preamplifier and scaler. A rate meter may also be useful for rapid selection of recording characteristics. Binary scalers are preferable to decade because they provide a finer choice of scale factors. Mechanical recording methods generally use scale factors of 4 to 1, and higher, due to the limited stylus response speed and the limited response range of the recording method. Scale factors should increase in steps up to a ratio of perhaps 1,000 to 1 to allow for all levels of activity up to large therapeutic doses. The use of a discriminator or a single-channel analyzer is valuable in excluding scattered radiation and thus improving contrast in the final display.

4.5.6. Recording Systems

The recording method plays an important role in defining the limitations of scanning techniques. In addition to the conventional solenoid-operated punch method of recording, many techniques have been developed for giving improved visibility to small areas of increased or decreased count rate. One method is the "background erase" technique, in which the counts are recorded only if the count rate exceeds a preset level. With this method great care is needed to avoid being misled by an unsuitable choice of the preset level. A variation of this technique involves the use of a "background subtract" circuit, whereby a uniform preset count rate is subtracted from the entire scan pattern. In photoscanning, the counter signal actuates a flashing light to which photographic film is exposed. If conditions are properly chosen so that a modest increase in the original signal produces a great increase in the density on the photographic film, the contrast of the original record is enhanced on the photoscan. Photocopying methods may also be used to erase background and increase contrast on scan recordings without destroying the original recording. One scanner has been developed using a multi-colored ribbon between the solenoid operated punch and the recording paper, the color selection depending on count rate.

All of these methods involve loss of some of the information originally received by the detector. The best use of the equipment is achieved in systems such as that in which each impulse is stored on magnetic tape. This can then be played back with various levels of background suppression or contrast enhancement until optimum conditions are found.

4.6. Whole-Body Counting

4.6.1. Introduction

This procedure has so far found two main applications, (a) the identification and quantitative estimation of radionuclides present in the body and (b) the study of absorption and retention in the body of intentionally administered radionuclides or labelled compounds (Radioactivity in Man, 1961; Whole Body Counting, IAEA, 1961; Spiers, 1957). The choice of the method of measurement and of the equipment depends on the application.

4.6.2. Methods of Measurement

Identification of an unknown radionuclide requires spectral analysis of the radiations emitted from the subject under study. These are best detected by a scintillation material with high spectral resolution, usually a sodium iodide crystal. Spectra so obtained are distorted by self-absorption and scattering, within the body tissues, of the gamma rays (or, in the case of beta-ray emitters, of the bremsstrahlung). The extent of this distortion depends on the location of the activity within the body and on the energy of the radiations emitted. The spectrum is also affected by the presence of other radionuclides in the body such as the naturally occurring K^{40} and the fallout component Cs^{137}-Ba^{137}.

Once the radionuclide is identified, quantitative estimation of the amount present requires calibration of the detecting device against known amounts of the radionuclide in question using a phantom, or of a short-lived radionuclide with similar metabolic and radiation characteristics as that administered to the subject (e.g., K^{42} has been used for calibration of K^{40} measurements and Cs^{132} is likely to be equally useful for calibration in Cs^{137}-Ba^{137} measurements). The volume and shape of the phantom should resemble that of the subject studied. Some attempts are being made to simulate uneven distribution of the radionuclide within

the body by using compartmentalized phantoms. A further reduction of the effects of uneven distribution can be achieved by placing both the subject and the phantom in such a counting position that the sensitivity of the detecting device is similar for nearly all parts of the body. This is attempted either by arranging a number of crystals symmetrically around the body (or even by completely surrounding it by a liquid scintillation material) or by placing the subject in an arc with a single crystal at its center.

Calibration of other radionuclides is complicated by the presence of K^{40} and Cs^{137}–Ba^{137} in the body. Their contribution to the counting rate depends on their amount, on body build, and on the energy of the radiations emitted by the radionuclides studied in cases where the characteristic spectra overlap. If corrections for the presence of K^{40} and Cs^{137} in the subject are made using calibrations obtained with phantoms, it is necessary to make some rather crude assumptions based on data obtained in a large number of normal subjects counted under identical conditions.

The detection and quantitative estimation of naturally occurring radionuclides in human subjects is particularly important at low levels and has been primarily responsible for the development of whole-body-counting techniques. In this type of study, for example the study of natural K^{40} body burdens, shielding is of paramount importance in order to achieve the lowest possible background. Shielding of the detector itself, leaving an aperture for viewing the patient, is not sufficient because the patient's body will cause some net background alterations. This alteration may be a reduction, or in some circumstances, an increase due to scattering with a resultant change in spectral distribution. A shielded room or enclosure constructed of low-activity materials is therefore very desirable.

In studies with intentionally administered radionuclides the situation is somewhat less difficult, since a "background" spectrum for the subject can be obtained before the radionuclide is given. In such studies it is usually desired to follow the retention of the radionuclide in the body and to express the amount retained as a fraction of the initial burden. However, during the first few hours or even days after administration, the pattern of distribution of the radionuclide may undergo considerable change due to repartition between different body compartments and to specific accumulation in certain tissues. Thus, due to variations in geometry and self-absorption, the spectrum changes, and the total counting rate may even increase despite an actual loss of radioactivity from the body. Unless very careful attention is paid to positioning the subject so as to minimize these effects, data for body retention may be meaningless.

4.6.3. Equipment Requirements

The amount of radioactivity involved in the measurement of body burdens is usually quite small and the sensitivity of the detecting device should therefore be high. Scintillation detectors are the instrument of choice. If high spectral resolution is also required to identify one or several unknown radionuclides, sodium iodide crystals are to be preferred. Some workers favor a single large crystal because resolution is better and calibration problems are perhaps less troublesome; others prefer multi-crystal systems, which are advantageous in permitting simultaneous partial body analysis (International Directory, IAEA, 1962).

Most shielding rooms or enclosures are constructed of lead-lined iron; other shielding materials such as talc, chalk, and other minerals with low K^{40}, thorium and radium content are also in use. Care should be taken to avoid the use of steel which, during its production, has been contaminated with Co^{60}. The counting room should be well ventilated with filtered air to maintain a low level of radon daughter products within the enclosed volume. Aged air from storage tanks has occasionally been used to reduce the level of radon daughters or of A^{41} given off by nearby gas-cooled reactors. The room must be kept scrupulously clean and it is helpful to line the surface with a low-activity synthetic plastic; subjects to be counted should be showered and scrubbed to remove surface contamination, and provided with monitored clothing. With small body burdens, counting times of 30–60 minutes may be required to arrive at reasonable counting statistics; claustrophobia of the subject may be a problem.

The spectrum obtained is recorded by a multichannel analyzer. Channel stability is most important. Fully transistorized instruments are now available which require little attention and can be coupled with various automatic devices, such as tape recorders, computers and data printers, which may greatly ease the task of processing the wealth of information obtained. If it is desired to subtract one spectrum from another automatically, special techniques may be needed to keep energy discrimination very stable; a gain shift of more than one-half percent might introduce gross errors in the analysis.

If the radionuclide present in the body is known, then good energy resolution is no longer essential and can be sacrificed in favor of sensitivity or simplicity. Several institutions use liquid-scintillation counters made in the form of a hollow cylinder which surrounds the body and to which a large number of multiplier phototubes is attached; a distinct advantage of this arrangement is the short counting period. The use of a multichannel analyzer is no longer required. (See figs. 9.4–1 and 9.4–2, NCRP–NBS Handbook 80, 1961.)

In metabolic work, knowledge of retention data alone is of little value unless supplemented by other measurements on rates of distribution or organ uptake. This usually requires blood-plasma-assay procedures, and hence the sensitivity of additional apparatus for small sample measure-

ment determines the amount of radionuclide to be administered. Thus the high sensitivity of the devices for whole-body counting described above may be of little advantage in metabolic work unless sample-counting equipment of adequate sensitivity and stability is provided. Simple whole-body counters can frequently be used, and useful results have already been obtained without an iron room, employing a single-channel analyzer only.

4.6.4. Some Applications

In monitoring radiation workers, whole-body counting is a useful technique for assaying contamination relative to the permissible body burden; likewise the effectiveness of various decontamination procedures can be ascertained with high sensitivity. On various occasions, routine surveys of workers have revealed the presence of minute amounts of such radionuclides as Zn^{65}, Cs^{137}, Pu^{239}, and others, to which the subjects have unknowingly been exposed under normal and, presumably, safe working conditions. Another application involves the assay of total body Na^{24} induced by accidental exposure of a subject to a neutron beam, although much information on the neutron dose can also be obtained by simply analyzing a plasma sample for its Na^{24} content.

The technique has also been used for estimation of contamination (with radium and its daughter products) of watch-dial painters, and other radium workers, as well as with thorium and its daughter products of patients who had received a colloidal compound of thorium for diagnostic purposes. Such measurements can be usefully combined with analyses of exhaled breath for radon or thoron content respectively. It has recently been found that various groups of dial painters have become contaminated with substantial amounts of Sr^{90}; these cases are at present being carefully studied and attempts are being made to use whole-body counting of bremsstrahlung to assay total body burdens.

Whole-body-counting techniques are growing in use in clinical research work. Here their main advantage lies in effectively replacing the troublesome and invariably incomplete collection of excreta over long periods of time. Na^{22}, Ca^{47}, Fe^{59}, Sr^{85}, I^{131}, I^{131}-labeled proteins and Vitamin B_{12}, labeled with various radioactive cobalt isotopes, have been used to study retention in the body.

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Appendix I

Standards and Measurement of Radioactivity for Radiological Use (ICRU, 1959)*

14. Absolute and Relative Measurements

14.1. *Introduction.*

In this report two types of measurement are considered. Firstly, the preparation by absolute measurement, of so called "absolute standards" of radioactive nuclides. Secondly, the preparation, by comparative methods of measurement, of standards which may be called either reference standards or working standards; these standards will frequently be prepared by the use of a "standard instrument," calibrated by means of an absolute standard.

An absolute method is one in which a fundamental quantity such as rate of disintegration or rate of emission of energy is measured. A relative method is one which depends on a direct standardization. Terms such as "primary" and "secondary" have been used in the past to describe such standards but the present report is based on a system of nomenclature which has recently been proposed.[33]

14.2. *Absolute measurements.*

In view of the comprehensive review articles and detailed technical papers [31] which are available on absolute methods of measurement, a brief survey only is deemed necessary for the purpose of this report. A list of the methods applicable to nuclides used for radiological purposes is given in table 14.1.

The absolute measurement of the activity of a radioactive nuclide involves either the direct measurement of the rate R at which the radioactive nuclei in the sample are disintegrating or of the determination of the number N of unstable nuclei present in the sample and the decay con-

*Reprinted from National Bureau of Standards Handbook 78, Report of the International Commission on Radiological Units and Measurements (ICRU) 1959. (The section numbering is that of the original report.)

[31] A bibliography is appended.

[33] A bibliography is appended.

TABLE 14.1. *Absolute methods for the preparation of standards of radioactive nuclides*

Method	Nuclides to which method is applicable	Activity of measured source	Special corrections, limitations, etc.	Accuracy under most favorable conditions*
1. 4π *Counting*				
(a) Geiger-Müller counter	α emitters, β emitters, with or without γ rays.	0.01μc or less	Accuracy usually limited by uncertainty in corrections for loss of β particles in source material and source mount, particularly for β particle energies <0.5 Mev.	±2%
(b) Proportional counter	α emitters, β emitters, with or without γ rays.	0.1μc or less	Accuracy usually limited by uncertainty in corrections for loss of β particles in source material and source mount, particularly for β particle energies <0.5 Mev.	±2%
(c) High pressure proportional counter.	Electron-capturing nuclides	0.1μc or less	Method assumes knowledge of K-fluorescence yield and K/L capture ratio.	±5%
(d) Scintillation counting; solid phosphor.	β emitters, with or without γ rays.	0.1μc or less	Suitable for medium and high energy β emitters; low energy cut-off limits accuracy for low energy β emitters.	±2%
(e) Scintillation counting; liquid phosphor.	β emitters, with or without γ rays, α emitters.	0.1μc or less	Suitable for medium and high energy β emitters; low energy cut-off limits accuracy for low energy β emitters.	±2%
2. *Defined Solid Angle*				
(a) End window Geiger-Müller counter.	β emitters	0.1μc or more	Complex corrections for absorption and scattering effects	±5%
(b) Proportional counter	α emitters	0.1μc or more		±0.5%
	β emitters	0.1μc or more		±5%
(c) Zinc sulphide screen	α emitters	0.1μc or more		±0.5%
3. *Coincidence Counting*				
(a) $\beta\gamma$	$\beta+\gamma$ emitters having simple decay schemes.	1μc or less	Uncertainty in the correction for the gamma sensitivity of the β detector usually limits accuracy.	±2%
(b) $4\pi\beta$-γ	Many $\beta+\gamma$ emitters	0.1μc or less	Corrections, in general, very small and hence method is more accurate than $\beta\gamma$ method.	±1%
(c) $\gamma\gamma$	So far chiefly used for Co^{60}		Requires careful adjustment of efficiencies of counters, and allowance for angular correlation of γ rays.	±1%
(d) $X\gamma$ and $4\pi x$-γ	Electron-capturing nuclides	0.5μc or more		±5%
4. *Internal gas counting*	Low-energy β emitters. Electron-capturing nuclides.	0.01μc or less	Limited to radionuclide samples having suitable gaseous form.	±2%
5. *Measurement of loss of charge*	β emitters	0.1mc	Application limited by experimental difficulties	±5%
6. *Calorimetry*	α emitters	1mc or more		±0.5%
	β emitters	1mc or more	Involves a knowledge of the mean energy of the β particles.	±2%
	$\beta+\gamma$ emitters	1mc or more	Involves a knowledge of the mean energy of the β particles, the quantum energies of the γ rays, the number of γ quanta of each energy per disintegration and the fraction of the total γ radiation absorbed in the calorimeter.	±5%
Ionization measurement of dose rate.	γ emitters having relatively simple γ-ray spectra.	1mc or more	Accuracy of disintegration rate determination limited by uncertainty in W (energy per ion pair) for air and in some cases by incomplete knowledge of γ-ray spectra.	±4%
	β emitters		Accuracy limited by uncertainty in W and in average energy of β particles.	±5%
8. *Weighing*	Radium and long-lived radionuclides.	10 mg	High chemical and radio-chemical purity of radionuclide or parent essential.	±0.2%†
9. *Mass spectrometer*	Has been used for H^3			±1% (routinely)

*The figures given in this column are estimates of the extreme limits of total error in the disintegration-rate value, and the errors which contribute to this total depend on various factors which are not the same for each nuclide; the limits given indicate the accuracy believed to be obtainable under the most favorable conditions presently available.

†Radium standards.

stant of the nuclide. R and N are related by the radioactive decay law $R=\lambda N$. When $R=3.700 \times 10^{10}$ per second, the quantity of radionuclide present in the sample is 1 curie.

The most common modes of decay of radioactive nuclei are those involving the emission of an α or β particle with or without associated gamma radiation, or capture of an electron from one of the atomic electron shells with emission of x rays characteristic of the daughter nucleus, again with or without associated gamma-ray emission from the daughter nucleus.

14.2.a. *Alpha-particle standards.* Standard α-particle sources have been prepared from Po^{210}, uranium oxide, Pu^{239} and Am^{241} and thin sources of these can be satisfactorily calibrated in a $2\pi\alpha$ proportional flow counter. Sources of Po^{210} can also be calibrated in terms of α-particle emission, by measuring their rate of energy emission microcalorimetrically, or by finding their total γ-ray emission. The α-particle emission is then obtained using the energy of the α-particles or the branching ratio respectively.

14.2.b. *4π Geiger-Müller or gas-flow proportional counters.* For the absolute measurement of the activities of β emitters the $4\pi\beta$ method, using GM or gas-flow proportional counters has been adopted for routine use by most standardizing organizations. The relatively simple techniques for preparing thin conducting films have almost eliminated the need for a correction for loss of β particles by absorption in the source mount.

Current experiments indicate that losses due to absorption in the source material can be reduced to a minimum by employing certain techniques in the preparation of solutions and sources, e.g., in the measurement of the activity of S^{35}, a weak β emitter ($E_{max}=0.167$ Mev), carrier-free solutions are used and sources are redissolved and dried in an ammonia atmosphere.

The activities of samples of electron-capturing nuclides can be measured by counting the x photons in a 4π-gas-proportional counter. The pressure in the counter is varied over a range from one-half to several atmospheres, and the rate corresponding to infinite pressure (i.e., complete absorption of the x rays) is derived by extrapolation: alternatively, the counter is operated at a pressure sufficiently high to ensure practically complete absorption. The derivation of disintegration rate from the counting rate involves a knowledge of the fluorescence yield of the daughter nucleus and a correction for the absorption of the x rays in the foil used to absorb the Auger electrons. Agreement between this method and the coincidence method referred to later is good.

14.2.c. *4π Scintillation counters.* Scintillation counting of β particles in a 4π geometry using either solid or liquid phosphors has been shown to give good agreement with gas proportional counting, and has also been used for α-particle emitters. However, this technique is not suitable for the absolute measurement of the activities of low energy β emitters because the detection efficiency is very low for β particles having energies below a few kilovolts, and these form an appreciable fraction of the total. Moreover the background of thermal noise pulses from the electron-multiplier phototube reduces the accuracy of measurement for β particles below 10 kev. This background can be reduced by cooling and by using two or more phototubes in coincidence, but the latter method also reduces the sensitivity. The method may however be applicable to electron-capturing nuclides of sufficiently high atomic number because of the great efficiency of phosphors for the detection of x rays.

14.2.d. *Low-geometry defined-solid-angle counting.* End-window counters subtending a well-defined solid angle at the source have been used to measure a known fraction of the β particles emitted by a source. Although it is possible with care to reduce the errors associated with the corrections for absorption and scattering, the method is now seldom used for the direct standardization of β emitters.

Low-geometry defined-solid-angle systems in which the detector is a zinc sulphide screen and phototube have been shown to be capable of high precision in measurements of α-particle emitters.

14.2.e. *Coincidence counting.* The $\beta\gamma$ coincidence method provides an alternative, or an auxiliary, to $4\pi\beta$ counting of nuclides for which the β radiation is accompanied by γ rays, and for which the decay schemes are relatively simple and well-known, e.g., Co^{60}. Two detectors are required, one for β particles and one for γ photons, and either GM counters, proportional counters or scintillation counters are used. The method is generally slow unless scintillation counters are used in one or both channels; short resolution times (of the order of 10^{-8} second) are desirable in the coincidence circuits to reduce the random-coincidence rate when fast counting rates are employed. The corrections involved in the coincidence method are very much reduced in magnitude when the β detector is a 4π counter as in the so-called $4\pi\beta$-γ coincidence technique. Restriction to simple decay schemes is then less stringent.

The $\gamma\gamma$ coincidence method is of limited application but has been successfully used for Co^{60}. $X\gamma$ coincidence counting is used for the absolute measurement of the activities of electron-capturing nuclides for which the decay is accompanied by γ radiation, and has been used to check the validity of the extrapolation method described above.

14.2.f. *Internal gas counting, loss-of-charge method, calorimetry.* The absolute measurement of the activities of low-energy β emitters and electron-capturing nuclides has been effected by internal gas counting; the more general use of this method is desirable, but limitations are imposed by the difficulty of preparation of suitable gases.

The loss-of-charge method, in which the number of charged particles emitted by a sample is determined by measuring the total charge lost by the

sample, is complicated in practice by secondary effects, and is not generally a suitable alternative to the direct counting methods.

Calorimetry has been used for the intercomparison of national radium standards and for the standardization of α emitters and of β emitters for which the average energy of β decay is known. It is also used in conjunction with other measurements to determine the mean energy of β emission, especially when this is low. Calorimetry of γ-ray activity involves a knowledge of the γ-ray spectra and of the absorption of the γ radiation in the calorimeter, and is restricted in application and accuracy.

14.2.g. *Dosimetry:* In the measurement of activities by γ-ray dosimetry the experimental part consists of the determination of the exposure dose rate at a convenient distance from the sample; the disintegration rate is then obtained by dividing the dose rate at 1 cm, derived from this measurement, by the calculated value of the specific γ-ray emission (r-cm^2/mc-h) of the nuclide (table 14.6). The accuracy is limited by uncertainties in the values of a number of quantities on which the calculated value of the specific γ-ray emission depends, e.g., the energy required to produce an ion pair in air, and any uncertainty in the γ-ray spectrum of the nuclide. (For this reason, the measurement of disintegration rate by one or more alternative methods is often combined with exposure-dose-rate measurements in order to obtain experimental values of specific γ-ray emission, thereby providing a reliable basis for the preparation of reference or working standards, particularly of radio nuclides having complex γ-ray spectra.)

For β emitters, disintegration rates have been determined from measurements of the ionization current in a cavity chamber lined with a medium containing the radioactive material, or in a parallel-plate extrapolation chamber, one plate of which is formed by a solution of the material. The conversion from ionization current to disintegration rate involves the mean energy of β decay (table 14.7.) and the average energy required to produce an ion pair. (Conversely, one of these two quantities can be determined if the other is known and a sample of known disintegration rate is used.)

14.2.h. *Radiochemical purity.* Because of possible changes in radiochemical composition, it is preferable not to use short-lived standards for long periods of time, even though the half-life of the nuclide in question may be accurately known. Even in freshly-prepared standards unknown amounts of other isotopes may be present, e.g., P^{33} in P^{32}. It would be desirable for known radioactive contaminants to be specified by the supplier.

14.3. *International comparisons of standards.*

A list of the national laboratories participating in comparisons of measurements of standards is given in table 14.2. together with the methods used. Standards have been distributed for this purpose (column 2 table 14.2.) by the NBS[37] (C^{14}, Na^{24}, P^{33}, K^{42}, Co^{60}, Zn^{65}, $Sr^{90}+Y^{90}$, I^{131}, Au^{198}, Radium-226); the NPL (Na^{24}, P^{32}, Co^{60}, $Sr^{90}+Y^{90}$, I^{131}, Au^{198}); the AECL (H^3, S^{35}, P^{32}) and the CEA (S^{35}). The standards were ampoules containing a few milliliters of solution of the radionuclide; the activities ranged from a few microcuries to a few tens of microcuries per milliliter of solution.

TABLE 14.2—*National laboratories participating in comparisons of standards, and methods of measurement used*

Laboratory	Laboratories from which standards were received	Methods used by the laboratory (column 1) in comparisons of measurements of standards
Atomic Energy of Canada Limited, Chalk River (AECL).	NBS, NPL	$4\pi\beta$(PC), $4\pi\beta$(PC)γ.
Atomic Energy Research Establishment, Harwell (AERE).	AECL, NBS	$4\pi\beta$(GM), $\beta\gamma$(GM), $4\pi\beta$(GM)γ(LS), $4\pi\beta$(PC), $4\pi I$-γ.
Institut für Angewandte Radioaktivität, Leipzig (IAR).	AECL, NPL	$4\pi\beta$(GM), 4π(PC), $\beta\gamma$(GM).
Commissariat a l'Énergie Atomique, Saclay (CEA).	NBS, NPL	$4\pi\beta$(GM), $4\pi\beta$(PC), $\beta\gamma$, DSA.
Fondation Curie, Paris (FC)	NBS	$4\pi\beta$(PC).
Kernreaktor Isotopen Laboratorium Karlsruhe (KIL).	NBS	$4\pi\beta$(PC).
National Bureau of Standards, Washington (NBS).	AECL, NPL	$4\pi\beta$(PC), $\beta\gamma$, $\gamma\gamma$, LS, calorimetric.
National Physical Laboratory, Teddington (NPL).	AECL, CEA, NBS.	$4\pi\beta$(PC), $4\pi\beta$(PC), $\beta\gamma$ $4\pi\beta$(SS).
National Physical Research Laboratory, Pretoria (NPRL).	AECL, NBS, NPL.	$4\pi\beta$(PC), $4\pi\beta$(LS), $4\pi\beta$(PC)γ, $4\pi\beta$(LS)γ.
National Research Council, Ottawa (NRC).	AECL, NBS	$4\pi\beta$(PC), $4\pi\beta$(PC)γ.
Max Planck Institute, Göttingen (MPI).	CEA, NBS, NPL.	$4\pi\beta$(GM), $4\pi\beta$(PC), $\beta\gamma$.
Physikalisch-Technische Bundesanstalt, Braunschweig (PTB).	NBS, NPL	$4\pi\beta$(GM), $4\pi\beta$(PC), $\beta\gamma$, $4\pi\beta$(PC)γ.

The symbol (GM) denotes Geiger-Müller, (PC) proportional gas-flow (LS) liquid scintillation and (SS) solid scintillation counter. DSA denotes defined-solid-angle.

The results of measurements of the standards by the different organizations generally agree within ± 2 percent; in some cases there is a spread of 5 percent in the results, and only occasionally are there wider differences. Brief statements on the comparisons are given below, and several are summarized in table 14.3.

Hydrogen-3

Samples of H^3 distributed by AECL were measured at a number of research laboratories in North America, including AECL and NBS and also by the University of Glasgow in 1953. Agreement to within about ± 2.5 percent was obtained.

Sodium-24

The NPL measured NBS standards on three occasions during 1955–57; NBS $4\pi\beta$ values were on the average about 1 percent greater than NPL values. In an intercomparison between NBS, AECL and NPL in September, 1957 the spread of the individual values was about 3 percent.

The MPI values for NPL Na^{24} standards on four occasions during 1956–57 are on the average percent greater than NPL values. NPL standards were measured by the MPI, PTB and NPRL in 1957, the spread in the results (excluding one value) being 3 percent. In 1958 the spread in the results of measurements of NPL standards

[37] Please see table 14.2 for listing of laboratory abbreviations.

TABLE 14.3.—*International comparisons of standards*

Nuclide	Dates of comparisons	Laboratory issuing standard	Participating laboratories	Spread in values
				%
Na^{24}	1955–57	NBS	NBS, NPL	2
	1957	NBS	AECL, NBS, NPL	3
	1956–57	NPL	NPL, MPI	2
	1957	NPL	MPI, PTB, NPL, NPRL	3
	1958	NPL	AECL, IAR, NBS, NPL, PTB	3
P^{32}	1955–56	NBS	NBS, NPL	2
	1957	NPL	AECL, CEA, MPI, NPL, NPRL, PTB.	3
	1958	NPL	IAR	1
S^{35}	1955–56	AECL	AECL, AERE, NBS, NPL	4
	1958	CEA	CEA, MPI	2
K^{42}	1956–57	NBS	AECL, NBS, NPL	2
Co^{60}	1957	NPL	MPI, NPL	2.5
	1958	NPL	IAR, NPL	1.8
	1958	NPL	NPL, PTB	1
Zn^{65}	1958	NBS	AERE, NBS	1.5
Sr^{90}-Y^{90}	1957	NBS	NBS, NPL	0.6
	1957	NPL	NBS, NPL, MPI	2
	1958	NPL	IAR, NPL	1
	1958	NPL	NPL, PTB	1
I^{131}	1955–57	NBS	AECL, NBS, NPL	3
	1955	NPL	CEA, MPI, NPL	3.5
	1956–57	NPL	MPI, NPL	2
	1957	NPL	MPI, NPL, NPRL, PTB	4
	1958	NPL	AECL, NBS, MPI, NPL, PTB	3
Au^{198}	1955–56	NBS	AECL, NBS, NPL, NPRL	4
	1957	NPL	AECL, CEA, MPI, NPL, PTB.	4
	1958	NPL	NPL, PTB	2
	1958	NPL	IAR, NPL	5
	1958	NBS	AECL, CEA, FC, KIL, NBS, NPL, NPRL, NRC, PTB.	5

by AECL, NBS, NPL, PTB and IAR was also 3 percent.

Phosphorus-32

NPL measurements of three NBS P^{32} standards during 1955–56 agreed with NBS measurements to within ±1 percent, and measurements of NPL standards by the AECL, CEA, MPI, NPRL and PTB in 1957 agreed to within ±1.5 percent.

Sulfur-35

S^{35} standards were distributed by AECL for intercomparison in 1955. Results obtained by AERE, NBS, NPL, and the Royal Cancer Hospital agreed to within about ±2 percent of the AECL value. Results obtained by McGill University by $4\pi\beta$ counting were, however, some 10 percent higher than the AECL value. Measurements of CEA S^{35} standards by CEA and MPI in 1957 agreed to within 2 percent.

Potassium-42

Agreement within ±1 percent was obtained between measurements of NBS standards in 1956 and 1957 by AECL, NBS, and NPL.

Cobalt-60

A sample of a Co^{60} solution prepared by the NPL in 1955 and standardized by the $4\pi\beta$-γ coincidence method was measured by the MPI in 1957 by the $\beta\gamma$ coincidence method: the MPI value, after correction for decay, was 2.4 percent greater than the NPL value. A solution, also standardized at the NPL by the $4\pi\beta$-γ coincidence method in 1957 was measured by the IAR in 1958 by the $\beta\gamma$ coincidence method using GM counters; the IAR value was 1.8 percent greater than the NPL value. The PTB values by $4\pi\beta$-γ coincidence counting in 1958 were up to 1 percent greater than the NPL values.

Zinc-65

Measurements by AERE of NBS Zn^{65} standards in 1957 agreed with NBS measurements to within ±1.5 percent.

Strontium-Yttrium-90

Measurements by the NPL of NBS standards in 1957 agreed with NBS values to within ±0.3 percent. Measurements of NPL standards by the MPI and the NBS in 1957 and by the IAR and PTB in 1958 agreed with NPL values to within ±1 percent.

Iodine-131

The average of values obtained by the NPL by $4\pi\beta$ measurements of three NBS standards during 1955–57 was 2 percent less than the NBS average (which includes a correction of 2 percent for self-absorption); the AECL values in 1957 by the $4\pi\beta$-γ coincidence method were on the average 1.6 percent less than the NBS $4\pi\beta$ value (corrected for self-absorption).

In measurements of NPL standards during 1955–58, NBS $4\pi\beta$ values, with added absorption correction, and AECL $4\pi\beta$-γ values exceeded NPL values by about 2 percent, NPRL values by $4\pi\beta$-γ counting (using both liquid scintillation and proportional $4\pi\beta$ counters) exceeded NPL values by 1.5 percent, and CEA $4\pi\beta$ and $\beta\gamma$ values exceeded NPL values by about 3.5 percent. MPI and PTB values have agreed with NPL values to within ±1 percent.

A remarkably close gamma-ray analog for iodine-131, whose principal gamma rays have energies of 335 and 608 kev, has been produced by mixing the correct proportions of barium-133 (82-, 300-, and 375-kev gamma rays) and cesium-barium-137m (662-kev gamma rays). When such a simulated standard is used to calibrate a true iodine-131 sample, its effect as measured by any kind of gamma-sensitive equipment will be approximately the same as for iodine-131. It may thus have uses for calibration to within say ±3 percent when standards of short-lived iodine-131 itself do not happen to be available. A set of such "mock-iodine" standards consisting of two nominal 10-microcurie and two 50-microcurie ampoules was calibrated in the summer of 1955 at the MPI, NBS, and the NPL. The results, obtained by ionization chambers, Geiger-Müller and scintillation counters at these three laboratories did not differ by more than ±4 percent from the mean value.

Gold-193

Except for results which are in doubt due to self-absorption, measurements during 1955–56 of

NBS standards by AECL and NPL on three occasions, and by NPRL on one, agreed to within ±2 percent. Measurements of NPL standards by AECL, CEA, MPI, and PTB in 1957–58 also agreed to within about the same limits, as did measurements of NBS standards by AECL, CEA, FC, KIL, NPL, NPRL, NRC, and PTB.

Radium-226

New radium solution standards for radon analysis containing 10^{-9} and 10^{-11}g of radium-226 in 100 ml of carrier solution have been recently prepared at the National Bureau of Standards. Measurements of the 10^{-9}g standards at PTB agreed to within 0.3 percent with the NBS certified values. NBS measurements of PTB standards containing 4×10^{-9}g of radium-226 in 10 ml of solution agreed to within 0.5 percent of the PTB certified values.

14.4. *Relative measurements: preparation of reference or working standards.*

The preparation of a reference or working standard involves the comparison of the activity of a sample of a radioactive nuclide with that of a sample of the same nuclide, the activity of the latter sample having been established by one or more absolute methods. The comparison may either be made directly, using any suitable measuring system or through the medium of a calibrated standard instrument. Identity of conditions (form, geometry, etc.) for sample and standard or a knowledge of the corrections allowing for departure from identity is necessary. In principle any equipment which detects ionizing radiations can be used for relative measurements. For all such instruments it is necessary to use a suitable, long-lived, reference source in order to check the stability of the instrument over long periods of time. Counting systems have a wide application, especially for low activities, but they generally require frequent re-calibration. Halogen counters, when used under reproducible geometrical conditions (e.g., annular-type liquid counters for β emitters, well-type counters for γ emitters, etc.) maintain their calibration to within ±1 percent over long periods (see also 15.3). Other types are more variable and all have limited operational life. Comparisons of low-energy β emitters can sometimes be carried out using liquid-scintillation, organic-solution, or radioactive-gas counting.

The long-term reproducibility and simple operational characteristics of ionization chambers and current-measuring apparatus has led to a preference for this type of equipment for relative measurements. An optical-bench system using a cavity chamber provides the simplest conditions for precise direct comparisons of γ-ray sources when it is important to allow accurately for differences in their dimensions and form; for such systems activities of at least 10 μc are required. Chambers of the re-entrant cylinder (or well) type, known colloquially as $4\pi\gamma$ chambers, have proved very satisfactory for samples emitting gamma rays (table 14.4.) as have chambers having a 2π geometry, or an approximation thereto, for relative measurement of β activity (table 14.5.). Chambers providing "$4\pi\gamma$" and "$2\pi\beta$" facilities in separate or in single units have also been used. Forty commercially available composite $2\pi\beta$–$4\pi\gamma$ chambers, based on a design of the NPL Advisory Committee on Radioactive Standards, were tested at the NPL and found to have calibration factors differing by not more than ±1 percent from the averages for standard γ-ray sources, and by not more than ±3 percent from the averages for β-emitting sources.

14.5. *Specific gamma-ray emission.*

According to the 1956 ICRU Report the specific gamma-ray emission is the exposure dose rate produced by the unfiltered gamma rays from a point source of a particular radionuclide at a defined distance, and the unit for this quantity is

TABLE 14.4.—*Particulars of "$4\pi\gamma$" chambers used at national laboratories for the relative standardization of samples of radioactive nuclides*

Laboratory	Main dimensions of chamber			Size of cavity		Background current (inclusive of leakage)	Activity equivalent to background	Current-measuring apparatus	Remarks
	Ht	Diam	Vol	Depth	Diam				
	cm	*cm*	*liter*	*cm*	*cm*	*amp*			
National Bureau of Standards (USA).	30	25	15	23	2.5	3.6×10^{-13}	0.3 μc Co^{60}	Lindemann-Ryerson electrometer (automatic null method with kilocycle time measurement).	Response not critically dependent on source position and length.
National Physical Laboratory (UK):									
A	18	16.5	4	13	6.8	2×10^{-14}	0.3 μc Co^{60} 0.5 μg Ra	Lindemann electrometer (null method).	Response not critically dependent on source length or diameter.
B[a]	18	16.5	4	12	6.6	$1–2\times10^{-14}$	0.3 μc Co^{60} 0.5 μg Ra	Lindemann electrometer (null method); also D.C. amplifier.	Available commercially (with calibration data).
Commissariat à l'Énergie Atomique:									
A	34	13	2.3	34	7.5	5×10^{-14}	0.5 μc Co^{60}	D.C. amplifier	Argon; 3 atmospheres. Range 2μc to 1c Ra.
B	36	13	2	30	8	5×10^{-14}	0.05 μg Ra	D.C. amplifier	Argon; 20 atmospheres. Range 0.2 μc to 1c Ra.
Atomic Energy of Canada Limited.	28.5	17.1	13	32	7.9	7×10^{-14}	0.02 μc Co^{60}	Vibrating-reed electrometer and strip-chart recorder.	Argon; 20 atmospheres. Response not critically dependent on source position.
Physikalisch-Technische Bundesanstalt.	30	25	14.5	19	3.4	4×10^{-14}	0.33 μg Ra	D.C. amplifier	Response not critically dependent on source position and length.

[a] Combined $\beta+\gamma$ chamber designed by National Physical Laboratory Advisory Committee on Radioactive Standards.

TABLE 14.5.—*Particulars of beta-ray ionization chambers used at national laboratories for secondary standardization of samples of radioactive nuclides*

Laboratory	Geometry	Form	Source	Background current (inclusive of leakage)	Activity equivalent to background	Current measuring apparatus
				amp		
National Bureau of Standards (USA).	2π internal source.....	Sphere 15-cm diam.....	Dried deposit on disk.	3×10^{-13}	0.001 μc.....	Lindemann-Ryerson electrometer (automatic null method with kilocycle time measurements).
National Physical Laboratory (UK):						
A	2π (approx.) external source under wire mesh window.	Cylinder 7.5-cm diam. Cylinder 3.7-cm deep. Alkathene walls lined with graphite. Axial central electrode.	Dried deposit on disk.	10^{-14}	0.003 μc.....	Lindemann electrometer (null method).
B[a]	$<2\pi$ external source under dural foil window.	Parallel plate.....	1 ml liquid in plastic dish.	10^{-14}	0.04 μc P^{32}...	Lindemann electrometer (null method); also D.C. amplifier.
Commissariat à l'Énergie Atomique[b].	$<2\pi$ external source.....	Parallel plate.....	1 ml liquid in cylindrical vessel.	10^{-14}	1 μc P^{32}..... 10 μc Co^{60}	D.C. amplifier.

[a] Combined $\beta+\gamma$ chamber designed by National Physical Laboratory Advisory Committee on Radioactive Standards.
[b] Combined $\beta+\gamma$ chamber.

roentgens per millicurie hour at 1 cm; this unit has dimensions r-cm²/mc-h.

Theoretical and experimental values of the specific γ-ray emission (r/mch at 1 cm, or r-mc²/mc-h) are given in table 14.6. It should be noted that the calculated values do not include any contribution to the exposure dose rate made by x rays, e.g., bremsstrahlung or x rays following internal conversion or electron capture.

The experimental values given in the final column have been derived from measurements of the disintegration rates and exposure dose rates for samples of the radionuclides; the dose rates were measured using chambers calibrated with radium sources taking the specific γ-ray emission, Γ_{Ra}, for a source filtered with 0.5 mm platinum as 8.25 r/mgh at 1 cm. (The values should be multiplied by $\Gamma'_{Ra}/8.25$ if Γ_{Ra} is changed from 8.25 to Γ'_{Ra} r/mgh at 1 cm.) Consideration of the methods used in measuring exposure dose rates indicates that reasonable agreement may be expected between theoretical and experimental values for radionuclides such as Co^{60} for which the exposure dose rate is produced almost entirely by high energy γ radiation. However, for radionuclides such as Tm^{170} for which the γ radiation is of low energy and the exposure dose rate due to x rays may be appreciable and depend markedly on the form of the source, calculated values of the specific γ-ray emission must be used with caution in any radiological application. For such applications of these nuclides it is advisable to make a direct determination of the exposure-dose rate for a particular source.

18. References For Appendix I [40]

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4π *COUNTING*

a. b.[40a] *Geiger-Müller and Proportional*

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[40] References listed under subjects and in chronological order.

[40a] See table 14.1.

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TABLE 14.6.—*Specific gamma-ray emission*

Nuclide	Quantum energy in Mev and probability of emission		Specific gamma-ray emission, Γ (r-cm²/mc-h)	
	Annihilation	Nuclear gamma radiation	Theoretical [a]	Experimental [b]
Na^{22}	0.511(1.8)	1.28 (1)	11.9$_5$	
Na^{24}		1.37 (1)		18.7 [c]
		2.75 (1)	18.2$_3$	
K^{42}		1.53 (0.2)	1.3$_4$	1.4 [c]
Mn^{56}	.511(0.86)	0.73 (1)		
		0.94 (1)	18.5$_7$	
		1.45 (1)		
Mn^{54}		0.84 (1)	4.7$_0$	
Co^{58}	.511(0.3)	0.81 (1)	5.4$_4$	
Fe^{59}		0.191 (0.03) [d]	6.2$_1$	6.8 [c]
		1.096 (0.57)		
		1.289 (0.43)		
Co^{60}		1.173 (1)	12.9$_8$	13.1 [c]
		1.332 (1)		
Cu^{64}	.511(0.38)	1.34 (0.01)	1.1$_9$	
Zn^{65}	.511(0.034)	1.11 (0.49)	2.9$_9$	
Br^{82}		0.25 (0.06) [f]	14.6$_4$	15 [c]
		0.35 (0.03)		
		0.554 (0.75)		
		0.619 (0.42)		
		0.698 (0.28)		
		0.777 (0.83)		
		0.827 (0.25)		
		1.044 (0.28)		
		1.317 (0.27)		
		1.475 (0.17)		
Rb^{86}		1.08 (0.085) [g]	0.4$_0$	0.50 [h]
Mo^{99}		Many lines		1.29 [i]
Sb^{124}		0.60 (0.98)	9.8$_9$	
		0.64 (0.07)		
		0.72 (0.14)		
		0.97 (0.03)		
		1.06 (0.02)		
		1.33 (0.02)		
		1.37 (0.11)		
		1.45 (0.02)		
		1.61 (0.46)		
		2.11 (0.07)		
I^{126}		0.41 (0.23)	12.1$_3$	
		0.53 (1.0)		
		0.66 (1.0)		
		0.74 (0.99)		
		1.15 (0.31)		
I^{131}		0.08 (0.02)	2.2$_9$	2.23 [c]
		0.28 (0.08)		
		0.36 (0.80)		
		0.51 (0.01)		
		0.64 (0.09)		
		0.72 (0.03)		
I^{132}		2.2 (0.02)	12.3$_1$	12.0 [j]
		1.96 (0.04)		
		1.40 (0.13)		
		1.16 (0.10)		
		0.95 (0.23)		
		0.78 (0.18)		
		0.67 (1)		
		0.62 (0.06)		
		0.53 (0.18)		
$Cs^{137}+Ba^{137m}$		0.66 (0.81) [e]	3.0$_8$	3.4 [k]
Tm^{170}		0.084 (0.03) [e]	0.01$_1$	0.04 [l]
Ir^{192}		Many lines		5.0 [c]
Au^{198}		0.41 (0.96)	2.2$_2$	2.32 [c]
		0.68 (0.01)		
Ra^{226}+decay products (0.5 mm Pt filter).		Many lines		8.25 [l]

[a] For all the nuclides, with the exception of those based on reference (d), (f) and (g), the theoretical values are based on nuclear data supplied by the U.S. National Research Council Nuclear Data Group to the U.S. National Committee on Radiation Protection and Measurements for publication in the NCRP Manual on Radioactivity Procedures. The values of Γ have been calculated using a dose-rate versus photon-energy curve calculated by K. W. Geiger from the data given in table 8.1. for a value of W=34.0 ev per ion pair. The initial preparation of this table was carried out by N. G. Trott and Miss F. S. Lawson and the final calculation of Γ by W. B. Mann.

[b] The references given do not cover all published work.

[c] Perry, W. E. and Dale, J. W. G. (National Physical Laboratory) unpublished. The values are based on the specific gamma-ray emission of radium which has been taken to be 8.25 r-cm²/mc-h.

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Appendix II

Radiation Quantities and Units*

International Commission on Radiological Units and Measurements (ICRU) Report 10a 1962

1. Introduction

There has recently been much discussion of the fundamental concepts and quantities employed in radiation dosimetry. This has arisen partly from the rapid increase in the number of individuals using these concepts in the expanding field of nuclear science and technology, partly because of the need for extending the concepts so that they would be of use at higher photon energies and for particulate as well as for photon radiation, but chiefly because of certain obscurities in the existing formulation of the quantities and units themselves.

The roentgen, for example, was originally defined to provide the best quantitative measure of exposure to medium-energy x radiation which the measuring techniques of that day (1928) permitted. The choice of air as a standard substance was not only convenient but, also appropriate for a physical quantity which was to be correlated with the biological effect of x rays, since the effective atomic number of air is not very different from that of tissue. Thus a given biological response could be reproduced approximately by an equal exposure in roentgens for x-ray energies available at that time. Since 1928 the definition of the roentgen has been changed several times, and this has reflected some feeling of dissatisfaction with the lack of clarity of the concept.

The most serious source of confusion was the failure to define adequately the radiation quantity of which the roentgen was said to be the unit.[1] As a consequence of this omission the roentgen had gradually acquired a double role. The use of this name for the unit had become recognized as a way of specifying not only the magnitude but also the nature of the quantity measured. This practice conflicts with the general usage in physics, which permits, within the same field, the use of a particular unit for all quantities having the same dimensions.

Even before this, the need for accurate dosimetry of neutrons and of charged particles from accelerators or from radionuclides had compelled the International Commission on Radiological Units and Measurements (ICRU) to extend the number of concepts. It was also desired to introduce a new quantity which could be more directly correlated with the local biological and chemical effects of radiation. This quantity, *absorbed dose*, has a generality and simplicity which greatly facilitated its acceptance, and in a very few years it has become widely used in every branch of radiation dosimetry.

The introduction of absorbed dose into the medical and biological field was further assisted by defining a special unit—the *rad*. One rad is approximately equal to the absorbed dose delivered when soft tissue is exposed to 1 roentgen of medium voltage x radiation. Thus in many situations of interest to medical radiology, but not in all, the numbers of roentgens and rads associated with a particular medical or biological effect are approximately equal and experience with the earlier unit could be readily transferred to the new one. Although the *rad* is merely a convenient multiple of the fundamental unit, erg/g, it has already acquired, at least in some circles, the additional connotation that the only quantity which can be measured in rads is absorbed dose. On the other hand, the rad has been used by some authors as a unit for a quantity called by them *first collision dose;* this practice is deprecated by the Commission.

Being aware of the need for preventing the emergence of different interpretations of the same quantity, or the introduction of undesirable, unrelated quantities or units in this or similar fields of measurement, the ICRU set up, during its meeting in Geneva in September 1958, an *Ad Hoc* Committee. The task of this committee was to review the fundamental concepts, quantities, and units which are required in radiation dosimetry and to recommend a system of concepts and a set of definitions which would be, as far as possible, internally consistent and of sufficient generality to cover present requirements and such future requirements as can be foreseen. The committee was instructed to pay more attention to consistency and rigor than to the historical development of the subject and was authorized to reject any existing quantities or units which seemed to hinder a consistent and unified formulation of the concepts.

Bertrand Russell [2] in commenting on the use and abuse of the concept of infinitesimals by mathematicians, remarks: "But mathematicians did not at first pay heed to (these) warnings. They went ahead and developed their science, and it is well that they should have done so. It is a peculiar fact about the genesis and growth of new disciplines that too much rigor too early imposed stifles the imagination and stultifies invention. A certain freedom from the strictures of sustained formality tends to promote the development of a subject in its early stages, even if this means the risk of a certain amount of error. Nonetheless, there comes a time in the development of any field when standards of rigor have to be tightened."

The purpose of the present reexamination of the concepts to be employed in radiation dosimetry

*Reprinted from National Bureau of Standards Handbook 84, Radiation Quantities and Units, Internation Commission on Radiological Units and Measurements, ICRU Report 10a (1962). The Section numbering is that of the original report.

[1] Fränz, H. and Hübner, W. Concepts and Measurement of Dose, Proceedings of Second International Conference on the Peaceful Uses of Atomic Energy, Geneva 1958, P/971 21, 101, United Nations, Geneva (1958).

[2] Russell, B., Wisdom of the West, p. 280 (Doubleday and Co, Inc., New York 1959).

was primarily "to tighten standards of rigor". If in the process, some increased formality is required in the definitions in order to eliminate any foreseeable ambiguities, this must be accepted.

2. General Considerations

The development of the more unified presentations of quantities and units which is here proposed was stimulated and greatly assisted by mathematical models of the dosimetric field which had been proposed by some members of the committee in an effort to clarify the concepts. It appeared, however, that the essential features of the mathematical models had been incorporated into the definitions and hence the need for their exposition in this report largely disappeared. The mathematical approach is published elsewhere.[3]

As far as possible, the definitions of the various fundamental quantities given here conform to a common pattern. Complex quantities are defined in terms of the simpler quantities of which they are comprised.

The passage to a "macroscopic limit" which has to be used in defining point quantities in other fields of physics can be adapted to radiation quantities and a special discussion of this is included in the section headed "limiting procedures".

The general pattern adopted is to give a short definition and to indicate the precise meaning of any special phrase or term used by means of an explanatory note following the definition. There has been no attempt to make the list of quantities which are defined here comprehensive. Rather, the Commission has striven to clarify the fundamental dosimetric quantities and a few others (such as activity) which were specifically referred to it for discussion.

It is recognized that certain terms for which definitions are proposed here are of interest in other fields of science and that they are already variously defined elsewhere. The precise wording of the definition and even the name and symbol given to any such quantity, may at some future date require alteration if discussions with representatives of the other interested groups of scientists should lead to agreement on a common definition or symbol. Although the definitions presented here represent some degree of compromise, they are believed to meet the requirements in the field of radiation dosimetry.

3. Quantities, Units, and Their Names

The Commission is of the opinion that the definition of concepts and quantities is a fundamental matter and that the choice of units is of less importance. Ambiguity can best be avoided if the defined quantity which is being measured is specified. Nevertheless, the special units do exist in this as in many other fields. For example, the hertz is restricted, by established convention, to the measurement of vibrational frequency, and the curie, in the present recommendations, to the measurement of the activity of a quantity of a nuclide. One does not measure activity in hertz nor frequency in curies although these quantities have the same dimensions.

It was necessary to decide whether or not to extend the use of the special dosimetric units to other more recently defined quantities having the same dimensions, to retain the existing restriction on their use to one quantity each, or to abandon the special units altogether. The Commission considers that the addition of further special units in the field of radiation dosimetry is undesirable, but continues to recognize the existing special units. It sees no objection, however, to the expression of any defined quantity in the appropriate units of a coherent physical system. Thus, to express absorbed dose in ergs per gram or joules per kilogram, exposure in coulombs per kilogram or activity in reciprocal seconds, are entirely acceptable alternatives to the use of the special units which, for historical reasons, are usually associated with these quantities.

The ICRU recommends that the use of each special unit be restricted to one quantity as follows:

The rad—solely for absorbed dose
The roentgen—solely for exposure
The curie—solely for activity.

It recommends further that those who prefer to express quantities such as absorbed dose and kerma (see below) in the same units should use units of an internationally agreed coherent system.

Several new names are proposed in the present report. When the absorbed dose concept was adopted in 1953, the Commission recognized the need for a term to distinguish it from the quantity of which the roentgen is the unit. In 1956 the Commission proposed the term *exposure* for this latter quantity. To meet objections by the ICRP, a compromise term, "exposure dose" was agreed upon.[4] While this term has come into some use since then, it has never been considered as completely satisfactory. In the meantime, the basic cause of the ICRP objection has largely disappeared since most legal codes use either the units rad or rem.

Since in this report the whole system of radiological quantities and units has come under critical review, it seemed appropriate to reconsider the 1956 decision. Numerous names were examined as a replacement for exposure dose, but there were serious objections to any which included the word dose. There appeared to be a minimum of objection to the name *exposure* and hence this term has been adopted by the Commission with the hope that the question has been permanently settled. It involves a minimum change from the older name exposure dose. Furthermore, the elimination of the term "dose" accomplishes the long-felt desire of the Commission to retain the term dose for one quantity only—the absorbed dose.

[3] Rossi, H. H. and Roesch, W. C., Field Equations in Dosimetry, Radiation Res. 16, 783 (1962).

[4] For details see ICRU, 1956 Report, NBS Handb. 62, p. 2 (1957).

The term "*RBE* dose" has in past publications of the Commission not been included in the list of definitions but was merely presented as a "recognized symbol." In its 1959 report the Commission also expressed misgivings over the utilization of the same term, "*RBE*", in both radiobiology and radiation protection. It now recommends that the term *RBE* be used in radiobiology only and that another name be used for the linear-energy-transfer-dependent factor by which absorbed doses are to be multiplied to obtain, for purposes of radiation protection, a quantity that expresses, on a common scale for all ionizing radiations, the irradiation incurred by exposed persons. The name recommended for this factor is the *quality factor*, (*QF*). Provisions for other factors are also made. Thus a *distribution factor*, (*DF*), may be used to express the modification of biological effect due to non-uniform distribution of internally deposited isotopes. The product of absorbed dose and modifying factors is termed the *dose equivalent*, (*DE*). As a result of discussions between ICRU and ICRP the following formulation has been agreed upon:

The Dose Equivalent

1. For protection purposes it is useful to define a quantity which will be termed the "dose equivalent", (*DE*).
2. (*DE*) is defined as the product of absorbed dose, *D*, quality factor, (*QF*), dose distribution factor, (*DF*), and other necessary modifying factors.

$$(DE) = D\ (QF)\ (DF) \ldots .$$

3. The unit of dose equivalent is the "rem". The dose equivalent is numerically equal to the dose in rads multiplied by the appropriate modifying factors.

Although this statement does not cover a number of theoretical aspects (in particular the physical dimensions of some of the quantities) it fulfills the immediate requirement for an unequivocal specification of a scale that may be used for numerical expression in radiation protection.

Another new name is that for the quantity which represents the kinetic energy transferred to charged particles by the uncharged particles per unit mass of the irradiated medium. This is the same as one of the common interpretations of a concept "first collision dose," that has proved to be of great value in the dosimetry of fast neutrons. The concept is also closely related to the energy equivalent of exposure in an x-ray beam. The name proposed, *kerma*, is based on the initials of *k*inetic *e*nergy *r*eleased in *ma*terial.

Still another new name is the *energy fluence* which is here attached to the quantity in the 1953 ICRU report called *quantity of radiation*. The latter term was dropped in the 1956 ICRU report but the concept—time integral of intensity—remains a useful one and the proposed term appears to be acceptable in other languages as well as English. A related quantity, *particle fluence*, which is equivalent to the quantity *nvt* used in neutron physics, is included to round out the system of radiation quantities.

The quantity for which the curie is the unit was referred to the committee for a name and definition. Hitherto the curie has been defined as a *quantity of the radioactive nuclide* such that 3.7×10^{10} disintegrations per second occur in it. However, it has never been specified what was meant by quantity of a nuclide, whether it be a number, mass, volume, etc. Meanwhile the custom has grown of identifying the number of curies of a radionuclide with its transformation rate. Because of the vagueness of the original concept, because of the custom of identifying curies with transformation rate and because it appeared not to interfere with any other use of the curie, the Commission recommends that the term *activity* be used for the transformation rate, and that the curie be made its unit. It is recognized that the definition of the curie is of interest to other bodies in addition to the ICRU, but by this report we recommend that steps be taken to redefine it as $3.7\times10^{10}\text{s}^{-1}$, i.e., as a unit of activity and not of quantity of a radioactive nuclide.

It is also recommended that the term *specific gamma-ray constant* be used instead of *specific gamma-ray emission* for the quotient of the exposure rate at a given distance by the activity. The former term focuses attention on the *constancy* of this quotient for a given radio nuclide rather than the *emission* of the source.

4. Detailed Considerations

A. Limiting Procedures

Except in the case of a uniform distribution of sources throughout a large region, radiation fields are in general non-uniform in space. They may also be variable in time. Many of the quantities defined in this report have to be specified as functions of space or time, and in principle they must therefore be determined for sufficiently small regions of space or intervals of time by some limiting procedure. There are conceptual difficulties in taking such limits for quantities which depend upon the discrete interactions between radiations and atoms. Similar difficulties arise with other macroscopic physical quantities such as density or temperature and they must be overcome by means of an appropriate averaging procedure.

To illustrate this procedure we may consider the measurement of the macroscopic quantity "absorbed dose" in a non-uniform radiation field. In measuring this dose the quotient of energy by mass must be taken in an elementary volume in the medium which on the one hand is so small that a further reduction in its size would not appreciably change the measured value of the quotient energy by mass and on the other hand is still large enough to contain many interactions and be traversed by many particles.[5] If it is

[5] In interpreting radiation effects the macroscopic concept of absorbed dose may not be sufficient. Whenever the statistical fluctuations around the mean value are important, additional parameters describing the distribution of absorbed energy on a microscopic scale are necessary.

impossible to find a mass such that both these conditions are met, the dose cannot be established directly in a single measurement. It can only be deduced from multiple measurements that involve extrapolation or averaging procedures. Similar considerations apply to some of the other concepts defined below. The symbol Δ precedes the symbols for quantities that may be concerned in such averaging procedures.

In the measurement of certain material constants such as stopping power, absorption coefficient, etc., the limiting procedure can be specified more rigorously. Such constants can be determined for a given material with any desired accuracy without difficulties from statistical fluctuations. In these cases the formula quoted in the definitions are presented as differential quotients.

B. Spectral Distributions and Mean Values

In practice many of the quantities defined below to characterize a radiation field and its interaction with matter are used for radiations having a complex energy spectrum. An important general concept in this connection is the *spectral concentration* of one quantity with respect to another. The spectral concentration is the ordinate of the distribution function of the first quantity with respect to the second. The independent quantity need not always be energy or frequency; one can speak of the spectral concentration of flux density with respect to quantum energy or of the absorbed dose with respect to linear energy transfer. The interaction constants (such as μ, S and W) referred to in this report are often mean values taken over the appropriate spectral distributions of the corresponding quantities.

C. Units

For any of the quantities defined below the appropriate unit of an internationally agreed coherent system can be used. In addition certain special units are reserved for special quantities:

the rad for absorbed dose
the roentgen for exposure
the curie for activity.

D. Definitions

(1) *Directly ionizing particles* are charged particles (electrons, protons, α-particles, etc.) having sufficient kinetic energy to produce ionization by collision.

(2) *Indirectly ionizing particles* are uncharged particles (neutrons, photons, etc.) which can liberate directly ionizing particles or can initiate a nuclear transformation.

(3) *Ionizing radiation* is any radiation consisting of directly or indirectly ionizing particles or a mixture of both.

(4) The *energy imparted* by ionizing radiation to the matter in a volume is the difference between the sum of the energies of all the directly and indirectly ionizing particles which have entered the volume and the sum of the energies of all those which have left it, minus the energy equivalent of any increase in rest mass that took place in nuclear or elementary particle reactions within the volume.

NOTES: (a) The above definition is intended to be exactly equivalent to the previous meanings given by the ICRU to "energy retained by matter and made locally available" or "energy which appears as ionization, excitation, or changes of chemical bond energies". The present formulation specifies what energy is to be included without requiring a lengthy, and possibly incomplete, catalogue of the different types of energy transfer.

(b) Ultimately, most of the energy imparted will be degraded and appear as heat. Some of it, however, may appear as a change in interatomic bond energies. Moreover, during the degradation process the energy will diffuse and the distribution of heat produced may be different from the distribution of imparted energy. For these reasons the energy imparted cannot always be equated with the heat produced.

(c) The quantity *energy imparted to matter* in a given volume is identical with the quantity often called *integral absorbed dose* in that volume.

(5) The *absorbed dose* (D) is the quotient of ΔE_D by Δm, where ΔE_D is the energy imparted by ionizing radiation to the matter in a volume element, Δm is the mass of the matter in that volume element and Δ has the meaning indicated in section 4.A.

$$D=\frac{\Delta E_D}{\Delta m}$$

The special unit of absorbed dose is the *rad*.

$$1 \text{ rad}=100 \text{ erg/g}=\frac{1}{100} \text{ J/kg}$$

NOTE: J is the abbreviation for Joule.

(6) The *absorbed dose rate* is the quotient of ΔD by Δt, where ΔD is the increment in absorbed dose in time Δt and Δ has the meaning indicated in section 4.A.

$$\text{Absorbed dose rate}=\frac{\Delta D}{\Delta t}$$

A special unit of absorbed dose rate is any quotient of the rad by a suitable unit of time (rad/d, rad/min, rad/h, etc.).

(7) The *particle fluence* [6] or *fluence* (Φ) of particles is the quotient of ΔN by Δa, where ΔN is the number of particles which enter a sphere [7] of cross-sectional area Δa and Δ has the meaning indicated in section 4.A.

$$\Phi=\frac{\Delta N}{\Delta a}$$

[6] This quantity is the same as the quantity, *nvt*, commonly used in neutron physics.

[7] This quantity is sometimes defined with reference to a plane of area Δa instead of a sphere of cross-sectional area Δa. The plane quantity is less useful for the present purposes and it will not be defined. The two quantities are equal for a unidirectional beam of particles perpendicularly incident upon the plane area.

(8) The *particle flux density* or *flux density* (φ) of particles is the quotient of $\Delta\varphi$ by Δt where $\Delta\varphi$ is the particle fluence in time Δt and Δ has the meaning indicated in section 4.A.

$$\varphi=\frac{\Delta\Phi}{\Delta t}$$

NOTE: This quantity may also be referred to as particle fluence rate.

(9) The *energy fluence* (F) of particles is the quotient of ΔE_r by Δa, where ΔE_r is the sum of the energies, exclusive of rest energies, of all the particles which enter a sphere [8] of cross-sectional area Δa and Δ has the meaning indicated in section 4.A.

$$F=\frac{\Delta E_r}{\Delta a}$$

(10) The *energy flux density* or *intensity* (I) is the quotient of ΔF by Δt where ΔF is the energy fluence in the time Δt and Δ has the meaning indicated in section 4.A.

$$I=\frac{\Delta F}{\Delta t}$$

NOTE: This quantity may also be referred to as energy fluence rate.

(11) The *kerma* [9] (K) is the quotient of ΔE_K by Δm, where ΔE_K is the sum of the initial kinetic energies of all the charged particles liberated by indirectly ionizing particles in a volume element of the specified material, Δm is the mass of the matter in that volume element and Δ has the meaning indicated in section 4.A.

$$K=\frac{\Delta E_K}{\Delta m}$$

NOTES: (a) Since ΔE_K is the sum of the initial kinetic energies of the charged particles liberated by the indirectly ionizing particles, it includes not only the kinetic energy these charged particles expend in collisions but also the energy they radiate in bremsstrahlung. The energy of any charged particles is also included when these are produced in secondary processes occurring within the volume element. Thus the energy of Auger electrons is part of ΔE_K.

(b) In actual measurements Δm should be so small that its introduction does not appreciably disturb the radiation field. This is particularly necessary if the medium for which kerma is determined is different from the ambient medium; if the disturbance is appreciable an appropriate correction must be applied.

(c) It may often be convenient to refer to a value of kerma or of kerma rate for a specified material in free space or at a point inside a different material. In such a case the value will be that which would be obtained if a small quantity of the specified material were placed at the point of interest. It is, however, permissible to make a statement such as: "The kerma for air at the point P inside a water phantom is . . ." recognizing that this is a shorthand version of the fuller description given above.

(d) A fundamental physical description of a radiation field is the intensity (energy flux density) at all relevant points. For the purpose of dosimetry, however, it may be convenient to describe the field of indirectly ionizing particles in terms of the kerma rate for a specified material. A suitable material would be air for electromagnetic radiation of moderate energies, tissue for all radiations in medicine or biology, or any relevant material for studies of radiation effects.

Kerma can also be a useful quantity in dosimetry when charged particle equilibrium exists at the position and in the material of interest, and bremsstrahlung losses are negligible. It is then equal to the absorbed dose at that point. In beams of x or gamma rays or neutrons, whose energies are moderately high, transient charged-particle equilibrium can occur; in this condition the kerma is just slightly less than the absorbed dose. At very high energies the difference becomes appreciable. In general, if the range of directly ionizing particles becomes comparable with the mean free path of the indirectly ionizing particles, no equilibrium will exist.

(12) The *kerma rate* is the quotient of ΔK by Δt, where ΔK is the increment in kerma in time Δt and Δ has the meaning indicated in section 4.A.

(13) The *exposure* (X) is the quotient of ΔQ by Δm, where ΔQ is the sum of the electrical charges on all the ions of one sign produced in air when all the electrons (negatrons and positrons), liberated by photons in a volume element of air whose mass is Δm, are completely stopped in air and Δ has the meaning indicated in section 4.A.

$$X=\frac{\Delta Q}{\Delta m}$$

The special unit of exposure is the roentgen (R).

$$1R=2.58\times10^{-4}\text{C/kg}$$ [10]

NOTES: (a) The words "charges on all the ions of one sign" should be interpreted in the mathematically absolute sense.

(b) The ionization arising from the absorption of bremsstrahlung emitted by the secondary electrons is not to be included in ΔQ. Except for this small difference, significant only at high energies, the exposure as defined above is the ionization equivalent of the kerma in air.

[8] See footnote 7.

[9] Various other methods of specifying a radiation field have been used; e.g., for a neutron source the "first collision dose" in a standard material at a specified point (see Introduction).

[10] This unit is numerically identical with the old one defined as 1 e.s.u. of charge per .001293 gram of air. C is the abbreviation for coulomb.

(c) With present techniques it is difficult to measure exposure when the photon energies involved lie above a few Mev or below a few kev.

(d) As in the case of kerma (4D(11), note (c)), it may often be convenient to refer to a value of exposure or of exposure rate in free space or at a point inside a material different from air. In such a case the value will be that which would be determined for a small quantity of air placed at the point of interest. It is, however, permissible to make a statement such as: "The exposure at the point P inside a water phantom is . . . ."

(14) The *exposure rate* is the quotient of ΔX by Δt, where ΔX is the increment in exposure in time Δt and Δ has the meaning indicated in section 4.A,

$$\text{Exposure rate}=\frac{\Delta X}{\Delta t}$$

A special unit of exposure rate is any quotient of the roentgen by a suitable unit of time (R/s, R/min, R/h, etc.).

(15) The *mass attenuation coefficient* $\left(\frac{\mu}{\rho}\right)$ of a material for indirectly ionizing particles is the quotient of dN by the product of ρ, N, and dl, where N is the number of particles incident normally upon a layer of thickness dl and density ρ, and dN is the number of particles that experience interactions in this layer.

$$\frac{\mu}{\rho}=\frac{1}{\rho N}\frac{dN}{dl}$$

NOTES: (a) The term "interactions" refers to processes whereby the energy or direction of the indirectly ionizing particles is altered.

(b) For x or gamma radiations

$$\frac{\mu}{\rho}=\frac{\tau}{\rho}+\frac{\sigma}{\rho}+\frac{\sigma_{\text{coh}}}{\rho}+\frac{\kappa}{\rho}$$

where $\frac{\tau}{\rho}$ is the mass photoelectric attenuation coefficient, $\frac{\sigma}{\rho}$ is the total Compton mass attenuation coefficient, $\frac{\sigma_{\text{coh}}}{\rho}$ is the mass attenuation coefficient for coherent scattering, and $\frac{\kappa}{\rho}$ is the pair-production mass attenuation coefficient.

(16) The *mass energy transfer coefficient* $\left(\frac{\mu_K}{\rho}\right)$ of a material for indirectly ionizing particles is the quotient of dE_K by the product of E, ρ, and dl, where E is the sum of the energies (excluding rest energies) of the indirectly ionizing particles incident normally upon a layer of thickness dl and density ρ, and dE_K is the sum of the kinetic energies of all the charged particles liberated in this layer.

$$\frac{\mu_K}{\rho}=\frac{1}{E\rho}\frac{dE_K}{dl}$$

NOTES: (a) The relation between fluence and kerma may be written as

$$K=F\frac{\mu_K}{\rho}$$

(b) For x or gamma rays of energy $h\nu$

$$\frac{\mu_K}{\rho}=\frac{\tau_a}{\rho}+\frac{\sigma_a}{\rho}+\frac{\kappa_a}{\rho}$$

where

$$\frac{\tau_a}{\rho}=\frac{\tau}{\rho}\left(1-\frac{\delta}{h\nu}\right)$$

$\left(\frac{\tau}{\rho}\right.$=the photoelectric mass attenuation coefficient, δ=average energy emitted as fluorescent radiation per photon absorbed.$\left.\right)$ and

$$\frac{\sigma_a}{\rho}=\frac{\sigma}{\rho}\frac{E_e}{h\nu}$$

$\left(\frac{\sigma}{\rho}\right.$=total Compton mass attenuation coefficient, E_e=average energy of the Compton electrons per scattered photon.$\left.\right)$ and

$$\frac{\kappa_a}{\rho}=\frac{\kappa}{\rho}\left(1-\frac{2mc^2}{h\nu}\right)$$

$\left(\frac{\kappa}{\rho}\right.$=mass attenuation coefficient for pair production, mc^2=rest energy of the electron.$\left.\right)$

(17) The *mass energy-absorption coefficient* $\left(\frac{\mu_{en}}{\rho}\right)$ of a material for indirectly ionizing particles is $\frac{\mu_K}{\rho}(1-G)$ where G is the proportion of the energy of secondary charged particles that is lost to bremsstrahlung in the material.

NOTES: (a) When the material is air, $\frac{\mu_{en}}{\rho}$ is proportional to the quotient of exposure by fluence.

(b) $\frac{\mu_K}{\rho}$ and $\frac{\mu_{en}}{\rho}$ do not differ appreciably unless the kinetic energies of the secondary particles are comparable with or larger than their rest energy.

(18) The *mass stopping power* $\left(\frac{S}{\rho}\right)$ of a material for charged particles is the quotient of dE_e by the product of dl and ρ, where dE_e is the average energy lost by a charged particle of specified energy in traversing a path length dl, and ρ is the density of the medium.

$$\frac{S}{\rho}=\frac{1}{\rho}\frac{dE_e}{dl}$$

NOTE: dE_s denotes energy lost due to ionization, electronic excitation and radiation. For some purposes it is desirable to consider stopping power with the exclusion of bremsstrahlung losses. In this case $\frac{S}{\rho}$ must be multiplied by an appropriate factor that is less than unity.

(19) The *linear energy transfer* (L) of charged particles in a medium is the quotient of dE_L by dl where dE_L is the average energy locally imparted to the medium by a charged particle of specified energy in traversing a distance of dl.

$$L=\frac{dE_L}{dl}$$

NOTES: (a) The term "locally imparted" may refer either to a maximum distance from the track or to a maximum value of discrete energy loss by the particle beyond which losses are no longer considered as local. In either case the limits chosen should be specified.

(b) The concept of linear energy transfer is different from that of stopping power. The former refers to energy imparted within a limited volume, the latter to loss of energy regardless of where this energy is absorbed.

(20) The *average energy* (W) *expended in a gas per ion pair formed* is the quotient of E by N_W, where N_W is the average number of ion pairs formed when a charged particle of initial energy E is completely stopped by the gas.

$$W=\frac{E}{N_W}$$

NOTES: (a) The ions arising from the absorption of bremsstrahlung emitted by the charged particles are not to be counted in N_W.

(b) In certain cases it may be necessary to consider the variation in W along the path of the particle, and a differential concept is then required, but is not specifically defined here.

(21) A *nuclide* is a species of atom having specified numbers of neutrons and protons in its nucleus.

(22) The *activity* (A) of a quantity of a radioactive nuclide is the quotient of ΔN by Δt where ΔN is the number of nuclear transformations which occur in this quantity in time Δt and Δ has the meaning indicated in section 4.A.

$$A=\frac{\Delta N}{\Delta t}$$

The special unit of activity is the curie (c).

$$1c=3.7\times10^{10}s^{-1} \text{ (exactly)}$$

NOTE: In accordance with the former definition of the curie as a unit of quantity of a radioactive nuclide, it was customary and correct to say: "Y curies of P–32 were administered . . . ." It is still permissible to make such statements rather than use the longer form which is now correct: "A quantity of P–32 was administered whose activity was Y curies."

(23) The *specific gamma-ray constant* (Γ) of a gamma-emitting nuclide is the quotient of $l^2\frac{\Delta X}{\Delta t}$ by A, where $\frac{\Delta X}{\Delta t}$ is the exposure rate at a distance l from a point source of this nuclide having an activity A and Δ has the meaning indicated in section 4.A.

$$\Gamma=\frac{l^2\Delta X}{A\Delta t}$$

Special units of specific gamma ray constant are $Rm^2h^{-1}c^{-1}$ or any convenient multiple of this.

NOTE: It is assumed that the attenuation in the source and along l is negligible. However, in the case of radium the value of Γ is determined for a filter thickness of 0.5 mm of platinum and in this case the special units are $Rm^2h^{-1}g^{-1}$ or any convenient multiple of this.

TABLE 4.1. *Table of quantities and units*

No.	Name	Symbol	Dimensions [a]	Units		
				MKSA	cgs	Special
4	Energy imparted (integral absorbed dose).		E	J	erg	g. rad.
5	Absorbed dose	D	EM^{-1}	J kg^{-1}	erg g^{-1}	rad.
6	Absorbed dose rate.		$EM^{-1}T^{-1}$	J kg^{-1}s^{-1}	erg g^{-1}s^{-1}	rad s^{-1}, etc.
7	Particle fluence or fluence.	Φ	L^{-2}	m^{-2}	cm^{-2}	
8	Particle flux density.	φ	$L^{-2}T^{-1}$	m^{-2}s^{-1}	cm^{-2}s^{-1}	
9	Energy fluence	F	EL^{-2}	J m^{-2}	erg cm^{-2}	
10	Energy flux density or intensity.	I	$EL^{-2}T^{-1}$	J m^{-2}s^{-1}	erg cm^{-2}s^{-1}	
11	Kerma	K	EM^{-1}	J kg^{-1}	erg g^{-1}	
12	Kerma rate		$EM^{-1}T^{-1}$	J kg^{-1}s^{-1}	erg g^{-1}s^{-1}	
13	Exposure	X	QM^{-1}	C kg^{-1}	esu g^{-1}	R (roentgen).
14	Exposure rate		$QM^{-1}T^{-1}$	C kg^{-1}s^{-1}	esu g^{-1}s^{-1}	Rs^{-1}, etc.
15	Mass attenuation coefficient.	$\frac{\mu}{\rho}$	L^2M^{-1}	m^{2}kg^{-1}	cm^{2}g^{-1}	
16	Mass energy transfer coefficient.	$\frac{\mu_K}{\rho}$	L^2M^{-1}	m^{2}kg^{-1}	cm^{2}g^{-1}	
17	Mass energy absorption coefficient.	$\frac{\mu_{en}}{\rho}$	L^2M^{-1}	m^{2}kg^{-1}	cm^{2}g^{-1}	
18	Mass stopping power.	$\frac{S}{\rho}$	EL^2M^{-1}	J m^{2}kg^{-1}	erg cm^{2}g^{-1}	
19	Linear energy transfer.	L	EL^{-1}	J m^{-1}	erg cm^{-1}	kev(μm)$^{-1}$
20	Average energy per ion pair.	W	E	J	erg	ev.
22	Activity	A	T^{-1}	s^{-1}	s^{-1}	c (curie).
23	Specific gamma-ray constant.	Γ	QL^2M^{-1}	Cm^{2}kg^{-1}	esu cm^{2}g^{-1}	Rm^{2}h^{-1}c^{-1}, etc.
	Dose equivalent.	DE				rem.

[a] It was desired to present only one set of dimensions for each quantity, a set that would be suitable in both the MKSA and electrostatic-cgs systems. To do this it was necessary to use a dimension Q, for the electrical charge, that is not a fundamental dimension in either system. In the MKSA system (fundamental dimensions M, L, T, I) Q represents the product IT; in the electrostatic-cgs system (M, L, T) it represents $M^{1/2}L^{3/2}T^{-1}$.

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